Experimental test of recollision effects in double ionization of magnesium by near-infrared circularly polarized fields

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We report experimental results on correlated double ionization of magnesium (Mg) by near-infrared (0.8- and 1.03-μm) circularly polarized laser fields. With 0.8 μm, we confirm the recollision interpretation of the observed "knee" structure of Mg [Gillen *et al.*, Phys. Rev. A **64**, 043413 (2001)], even though the experiments are performed in the multiphoton regime. Our experiments show that, even in the multiphoton regime, which is normally thought to be a purely quantum-mechanical territory, some ionization phenomena can still be understood classically.

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Nonsequential double ionization (NSDI) (for a recent review, see, e.g., Ref. [1]) has been considered to be one of the most intriguing phenomena in strong-field atomic physics which shows an exceptionally high degree of electron-electron correlation. The most prominent feature of NSDI observed in early experiments [2–6] with noble-gas atoms is a characteristic knee structure in the yield curve of doubly charged ions versus laser intensity due to the enormous enhancement of ionization probabilities. The detected yields of double ionization could be several orders of magnitude greater than the predicted yields from sequential ionization. It was found that the knee structure disappeared when the laser was switched from linear to circular polarization [7] and consistent with the interpretation of electron recollision [8–10].

However, a knee structure in Mg atoms irradiated by 0.8-μm circularly polarized (CP) fields was later reported [11], and it seems to be in conflict with the recollision scenario. This puzzle was tackled theoretically in Refs. [12–14], and it was demonstrated with classical simulations that recollision could, in fact, happen under the experimental parameters in Ref. [11]. Numerous theoretical investigations and predictions followed [15–23], yet most of these works solely relied on the same data of Mg [11], and there is still a lack of further experimental tests in the literature. Although there are some recent ionization experiments utilizing two-color CP [24,25] or elliptical polarized (EP) [26] fields, the parameters in these measurements with noble gases are drastically different from Ref. [11] in the sense that these recent works are significantly closer to the tunneling regime.

In fact, it is surprising that a classical model is actually applicable for the case of Mg where the measurement was performed well into the multiphoton regime (the largest value of the Keldysh parameter $\gamma \sim 2$), in contrast to noble-gas experiments which are typically performed in the tunneling regime. The ionization potential of Mg is 7.64 eV, and for 0.8-μm lasers with photon energy 1.55 eV, it needs merely five photons to ionize the first electron. Can the knee structure observed with Mg really be explained by such a classical process as recollision as predicted by the above-cited theoretical studies?

In the present paper, we experimentally investigate, in the near-infrared regime, the wavelength and polarization dependence of the knee structure in magnesium and compare with the classical simulations. Our results provide concrete evidence to support the recollision interpretation for the knee structure in Mg at 0.8 μm [11].

Our experiments use a 0.8-μm 80-fs Ti:sapphire amplifier system at 1-kHz repetition rate (Spectra Physics: Spitfire Ace). Ion yield measurements are performed at two different wavelengths: 0.8 and 1.03 μm. The 1.03-μm beam is from the signal output generated by a homebuilt potassium titanyle arsenate optical parametric amplifier [27] pumped by the 0.8-μm laser. The laser pulses are focused by a lens into a homebuilt time-of-flight spectrometer in which ionization of atoms occurred. The laser pulse energy is controlled by a half-wave plate followed by a polarizer. The laser ellipticity is controlled by a quarter-wave plate. An effusive source oven is mounted below the interaction region for producing a beam of Mg or Zn atoms. Laser intensities are calibrated by the $10U_p$ break in the field-free photoelectron distributions of noble gases irradiated by linearly polarized pulses. Typically, the intensity uncertainty is within 20% [28]. When making measurements with noble-gas atoms, the oven is off, and the gas is delivered into the chamber through a leak valve.

We compare the experimental results with the classical ensemble simulations, a widely used simulation method of obtaining physical insight into strong-field ionization processes [29–41]. It is especially useful for the present case of strong-field double ionization with elliptically polarized laser fields, which two-electron time-dependent Schrödinger

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equation simulations are extremely demanding, if not impossible. The general idea of the classical ensemble method is to simulate the strong-field ionization process with an ensemble of classically modeled atoms. Soft-core Coulomb potentials $V_{\rm ion} = -1/\sqrt{|\mathbf{r}|^2 + a^2}$ are used to avoid autoionization of the classical atoms in the absence of an external field [42,43]. Therefore, the choice of the soft-core parameter a depends on the ground-state energy (sum of the first and second ionization potentials) of the atom. The a values for Mg and Zn are, thus, chosen to be 3.0 and 2.4, respectively. The electron-electron repulsion is modeled by a softened Coulomb potential $V_{ee} = 1/\sqrt{|\mathbf{r}_1 - \mathbf{r}_1|^2 + b^2}$ with $b = 1$. The initial electron positions and momenta are randomized with the constraint that the total energy of the two electrons equals the atomic ground-state energy. The electrons are then propagated classically under the influence of the laser field and the potentials. A double-ionization event happens if the final energies of both electrons are positive. More details about this type of simulation can be found in the aforementioned references.

It has been understood theoretically [12,14] that, for recollisions to happen with elliptical or circular polarization, the electron must be emitted with an initial transverse velocity that counteracts the drift velocity from the additional (minor) polarization direction, which tends to pull the electron away transversely and diminishes recollision. This drift velocity is $v_d = \varepsilon F_0/\omega$, where ε is the laser ellipticity, F_0 is the laser field amplitude at the time of emission, and ω is the laser angular frequency. If F_0 is estimated using an overbarrier condition $F_0 \approx I_p^2/4$, one gets $v_d \approx \varepsilon I_p^2/4\omega$.

On the other hand, the electron is emitted with an initial transverse velocity distribution. The detailed shape of this distribution, denoted $p(v_\perp)$, depends on atomic and laser parameters. For example, strong-field approximation theory predicts a Gaussian shape $p(v_\perp) \sim e^{-v_\perp^2/\sigma^2}$ of width $\sigma^2 = F_0/\sqrt{2I_p}$ [44] in agreement with experiment [45]. Using the overbarrier condition for F_0 we may estimate $\sigma \approx 0.42 I_p^{3/4}$.

Whether recollision is important with elliptical or circular polarization can be estimated from the comparison between v_d and σ: If v_d is much larger than σ, then recollision is negligible; otherwise if v_d is comparable to or smaller than σ, then recollision can be substantial. The ratio,

$$R \equiv \frac{v_d}{\sigma} \approx \frac{\varepsilon I_p^{5/4}}{1.68\omega} \tag{1}$$

is a dimensionless quantity measuring the importance of recollision with elliptical or circular polarization: the larger the ratio R, the less important the recollision process. From Eq. (1), there are three relevant parameters, namely, the laser angular frequency ω (or, equivalently, the wavelength λ), the laser ellipticity ε, and the atomic ionization potential I_p. Recollision and NSDI prefer (a) smaller λ (larger ω); (b) smaller ε; (c) smaller I_p. These are the testable predictions made from the recollision mechanism.

Our experimental results agree with these three predictions. Figure 1 shows the influence of laser wavelength on ionization of Mg with CP laser fields. Panel (a) shows ion yields of Mg^+ and Mg^{2+} as a function of laser intensity for 0.8 μm. Our results are in reasonable agreement with those of Gillen *et al.* [11]. Theoretical predictions using the Perelomov-Popov-Terent'ev (PPT) formula [46] for single ionization and sequential double ionization (SDI) are shown by the solid and dashed lines, respectively. The Mg^{2+} data clearly show a knee structure, deviating from the PPT predictions at low intensities. The knee structure disappears as the laser wavelength increases to 1.03 μm as shown in panel (c) where the Mg^{2+} data agree very well with the PPT calculations assuming SDI. Classical ensemble simulations [panels (b) and (d)] are in qualitative agreement with the data, showing a clear knee structure at 0.8 μm and no knee structure at 1.03 μm.

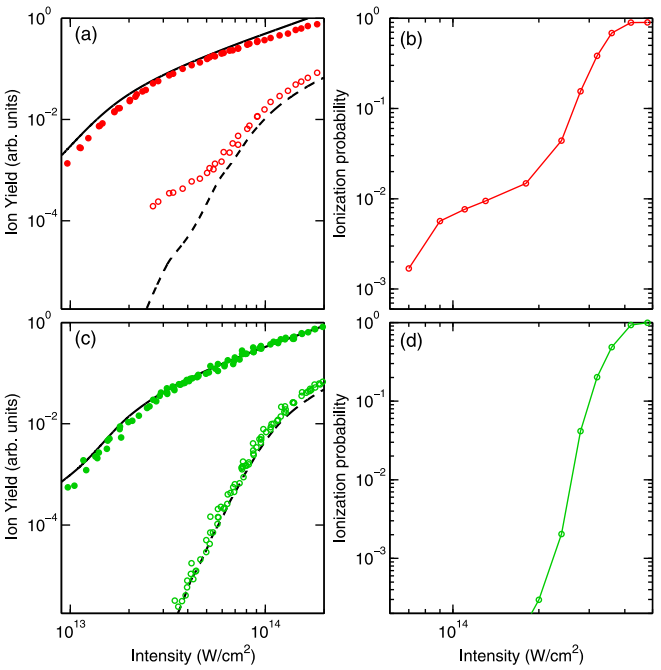

FIG. 1. Influence of wavelength: Ionization of Mg with CP laser fields at 0.8 μm (top row) and 1.03 μm (bottom row). The left column (a) and (c) shows experimental yields of Mg^+ (filled circles) and Mg^{2+} (open circles) as a function of laser intensity. Calculated yields using PPT formula for single and sequential double ionizations are shown by the solid and dashed lines, respectively. The right column (b) and (d) shows classical ensemble simulations for double ionization at the same two wavelengths.

Figure 2 shows the dependency of double ionization on laser ellipticity for Mg at the wavelength of 1.03 μm. Decreasing the ellipticity value from CP ($\varepsilon = 1$) to EP ($\varepsilon = 0.75$) leads to higher double-ionization yields, especially at low intensities. A weak but visible knee structure can be seen in the EP data below about 4×10^{13} W/cm^2 as indicated by the black arrow in Fig. 2(a). Classical ensemble simulations show qualitatively similar results as shown in Fig. 2(b).

Figure 3 shows the dependency of double ionization on the atomic ionization potential by comparing Mg with Zn, which has a (slightly) higher ionization potential of 9.39 eV. The knee structure shown with Mg disappears with Zn, consistent with the expectation from the recollision mechanism. Classical ensemble simulations are shown in Fig. 3(b). Although, in the simulations, the knee structure is not completely suppressed with Zn, it is much weaker than that with Mg. It is noteworthy that the absence of the knee structure was also observed in Ne and He at 0.61 μm [7], but these cases are

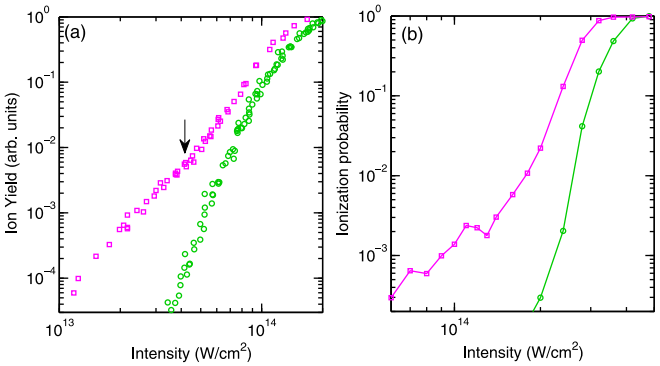

FIG. 2. Influence of ellipticity: Double ionization of Mg by CP and EP ($\varepsilon = 0.75$) fields at 1.03 μm. (a) Yields of Mg^{2+} for the case of CP (green circles) and EP (magenta squares) driving fields as a function of laser intensity. (b) Classical ensemble simulations for double-ionization probability with CP (green circles) and EP (magenta squares) fields.

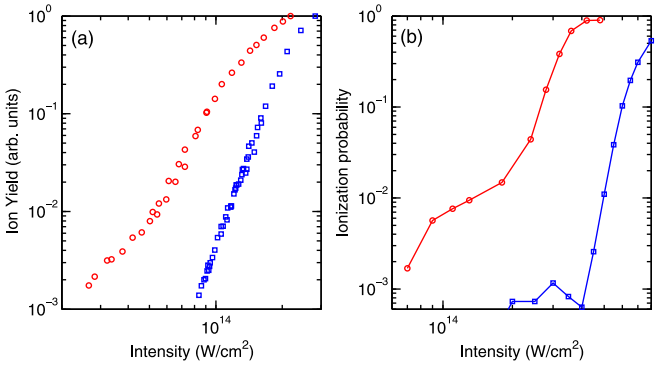

FIG. 3. Influence of I_p: Double ionization of Mg and Zn by 0.8-μm CP pulses. (a) Yields of Mg^{2+} (red circles) and Zn^{2+} (blue squares) as a function of laser intensity. For each atomic target, the yields are normalized with respect to the yield at the highest laser intensity in the dataset. (b) Classical ensemble simulations for double-ionization probability with Mg (red circles) and Zn (blue squares).

deep in the tunneling regime (for instance, $\gamma = 0.55$ for Ne at 10^{15} W/cm^2).

In conclusion, although the recollision interpretation of NSDI may appear, at first glance, inadequate in a multiphoton regime, it is confirmed by the experiment, in all its details (dependence on wavelength, ellipticity, and ionization potential). In addition, the recollision mechanism in CP has long been a surprise because the drift motion is easily thought as preventing recollisions. However, an initial momentum of the photoelectron is sufficient to generate trajectories that return to the parent ion [12], and it can be noted that the multiphoton regime of the first ionization is perfectly consistent with a substantial initial momentum. It could have been also conjectured that the spectrum of Mg energy levels, rich in doubly excited states, would prevent a classical trajectory interpretation. Our results show that this appears not to be the case. Studies in the near future will investigate the relation between quantum resonances and classical trajectories, especially at shorter wavelengths.

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