Unidirectional current excitation in tunneling ionization of asymmetric molecules

L. N. Alexandrov, M. Yu. Emelin,* and M. Yu. Ryabikin
Institute of Applied Physics, RAS, 46 Ulyanov Street, 603950, Nizhny Novgorod, Russia
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Asymmetric photocurrent produced by ionizing laser pulses in a gas is known to be a highly efficient source of terahertz radiation. We examine the possibility of exploiting the asymmetry of the medium itself, rather than the properties of the laser field acting on it, to facilitate the generation of directional photocurrents. We show that the magnitude of directional current and the efficiency of its excitation in tunneling ionization of asymmetric molecules can be significantly enhanced compared to the case of symmetric systems. The results obtained both in a simple classical model and in quantum-mechanical numerical simulations favor the subcycle asymmetry of the ionization process in combination with the effect of the Coulomb potential on the escaping electron as a mechanism responsible for a high-efficiency generation of residual current in tunneling ionization of oriented asymmetric molecules.

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I. INTRODUCTION

The widespread availability of compact femtosecond laser sources has greatly promoted the use of ultrashort pulses for the generation of terahertz pulses [1–3]. Recently, in studies on the interaction of intense femtosecond laser pulses with matter, much attention has been paid to the ionization-induced frequency conversion of laser pulses into low-frequency radiation, particularly into the terahertz range [4–13]. This transformation is closely related to the excitation of macroscopic quasi-dc currents [4–6,10]. Thus, in this approach, the efficiency of the generation of terahertz radiation is largely determined by the efficiency of directional current excitation.

The origin of the directional macroscopic current can be easily understood by the example of one-electron detachment. Assume for simplicity that at the ionization instant the electron velocity is zero and that after the detachment the electron motion is affected only by the laser field. If the ionization instant corresponds to the field maximum, the electron will acquire only an oscillatory component of the velocity, and after the laser pulse is over, the electron residual velocity will be equal to zero. Otherwise, if the ionization instant is phase shifted relative to the maximum of the field, the electron will acquire a nonzero drift velocity component, which will be preserved after the end of the pulse. If the average value of the drift velocity of the electrons released during the pulse will be nonzero, it would mean the creation of macroscopic residual current due to the laser pulse action on the sample. This current can lead to the generation of electromagnetic radiation whose frequency and excitation efficiency are determined by the geometry of the system, the density of the created plasma, and the value of the residual current.

From this mechanism, it follows that the key to the maximum directional current excitation is the optimal timing of ionization acts. As is known, in the case when the electron potential function of the gas particles is symmetric and the laser pulse is monochromatic, the final electron velocity distribution is symmetric; i.e., the macroscopic directional current is not created. To produce a current, it is necessary to break the symmetry of the ionization process. This can be done in several ways: (1) by using a few-cycle laser pulse [4,6,7,10], (2) by ionization with a two-color [5,8,9,11,12] or multicolor [13] field, and (3) by using an asymmetric potential. In this paper, we examine the last of these options. Although ionization of asymmetric molecules has recently attracted much interest (see, e.g., Refs. [14–16]), their potential use for the terahertz wave generation has not been addressed to date.

II. NUMERICAL SIMULATION

Study of the dependence of the residual current on the laser pulse parameters and on the properties of the gas molecules was carried out for a one-electron model of the molecular ion with a total charge of the nuclei equal to +3. The nuclear charge ratio ranged from 1:1 to 2:1 (the latter case corresponds to the HeH^{2+} molecular ion). The molecules were assumed to be oriented along the electric field vector. In experiment, field-free orientation of heteronuclear molecules can be induced by femtosecond two-color laser fields [17].

The quantum-mechanical treatment in this section is based on the numerical integration of the time-dependent Schrödinger equation (TDSE). To simplify the calculations, in the main part of them, instead of the three-dimensional potential, the one-dimensional model is used, in which the Coulomb factor $-1/r$ for each nucleus is replaced by the factor

$$f(x) = -(0.7 + x^2)^{-1/2} - 0.42(0.7 + x^2)^{-1}. \quad (1)$$

This factor has been chosen so that the dependence of the ionization probability on the internuclear distance and on the carrier-envelope phase (CEP) of the laser pulse for the one-dimensional (1D) model of the HeH^{2+} molecular ion would be close to that known for the three-dimensional case [18]. In the frozen-nuclei approximation, the TDSE for a one-electron diatomic molecular system interacting with a laser pulse can be written as

$$i\frac{\partial}{\partial t}\Psi(x,t) = \left[\frac{\hat{p}^2}{2} + A(t)\,\hat{p} + V(x)\right]\Psi(x,t), \quad (2)$$

*emelin@ufp.appl.sci-nnov.ru

where

$$V(x) = Z_1 f(x + D/2) + Z_2 f(x - D/2). \quad (3)$$

Here $\hat{p}$ is the electron momentum, Z_1 and Z_2 are the effective electric charges of the nuclei 1 and 2, respectively (hereafter we assume $Z_1 \geqslant Z_2$), D is the internuclear distance, and A is the vector potential of the linearly polarized laser field, which is defined as follows:

$$A(t) = A_0 \cos^2(\pi t/\tau) \sin(\omega t + \varphi), \quad -\frac{\tau}{2} < t < \frac{\tau}{2}. \quad (4)$$

Calculations were performed for a wide range of parameters. The laser pulse full width at zero level (τ) ranged from 1.5 to 81 cycles; the peak intensity of the laser pulse whose frequency was chosen to be the second harmonic of Ti:sapphire laser (wavelength 400 nm) was varied in the interval (3×10^{15}–2.5×10^{16}) W/cm^2. The internuclear distance (D) was regarded as a parameter, whose magnitude ranged from $1.6a_0$ to $5.6a_0$ (a_0 is the Bohr radius). The TDSE was integrated numerically using the fast-Fourier-transform-based split-operator technique [19].

The residual current observed after ionization of a molecule can be found as follows:

$$j_{\rm res}(D, Z_1/Z_2, A_0, \tau, \varphi) = \int_{-\tau/2}^{\tau/2} \ddot{d}(t)\, dt, \quad (5)$$

where $\ddot{d}$ is the second derivative of the dipole moment of the quantum system. As in Ref. [10], the residual current was normalized to the value of the oscillatory component of the current $j_{\rm osc} = E_0/\omega$ excited in the emerging plasma by the laser field of amplitude E_0 and frequency ω. The normalized residual current $j_{\rm norm} = j_{\rm res}/j_{\rm osc}$ does not depend on the gas density and characterizes the efficiency of excitation of plasma oscillations by the laser pulse. The numerical results are presented in Figs. 1–4.

Figure 1 shows the CEP dependence of the normalized residual current for various pulse durations, nuclear charge ratio 2:1, internuclear distance $3.4a_0$, and laser intensity 5×10^{15} W/cm^2. It is seen that, with increasing pulse duration, the current is less dependent on the CEP. At the same time, in contrast to the atomic case [10], the magnitude of the residual current remains large even for a rather long (nine-cycle) pulse. The leading role in creating the directional current in the long-pulse case is played by the asymmetry of the molecular potential (see below).

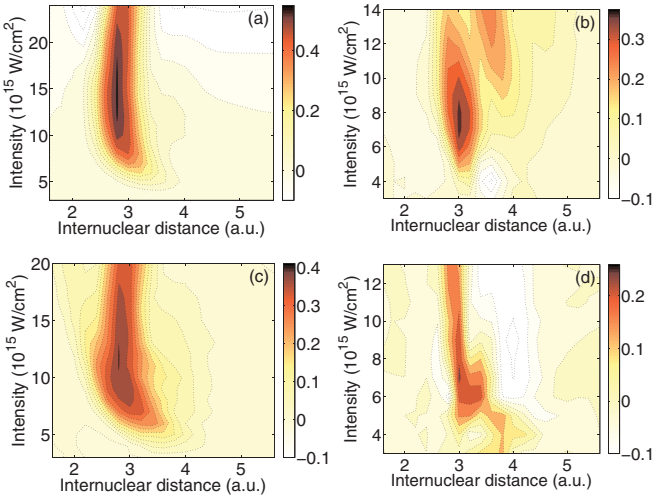

FIG. 2. (Color online) Normalized residual current as a function of laser intensity and internuclear distance for the cases of (a, c) an asymmetric molecule (charge ratio, 2:1) and (b, d) a symmetric molecule (charge ratio, 1:1). Laser pulse duration is as follows: (a, b) τ = three cycles and (c, d) τ = nine cycles.

Figure 2 plots the normalized residual current as a function of laser intensity and internuclear distance for three- and nine-cycle laser pulses and the different degrees of asymmetry of the molecular potential. The CEP is optimized for each intensity.

For an asymmetric molecule with a nuclear charge ratio of 2:1, the optimal internuclear distance is $2.8a_0$. The optimal laser intensity is 1.5×10^{16} and 1.2×10^{16} W/cm^2 for the pulse duration equal to three and nine cycles, respectively. The maximum value of the normalized residual current for these pulse durations is 0.56 and 0.39, respectively. The conversion efficiency of the laser pulse energy into the directional motion of the electrons can be estimated as the square of the normalized residual current and reaches up to 30% for a three-cycle pulse if the other parameters are optimal.

For a symmetric molecule, the optimal internuclear distance is $3a_0$. The optimal laser intensity is 8×10^{15} and 7×10^{15} W/cm^2 for three- and nine-cycle pulses, respectively. The maximum value of the normalized residual current for these pulse durations is 0.37 and 0.25, respectively. In this case, the efficiency of excitation of residual current does not exceed 13%; i.e., it is significantly lower than for asymmetric molecules.

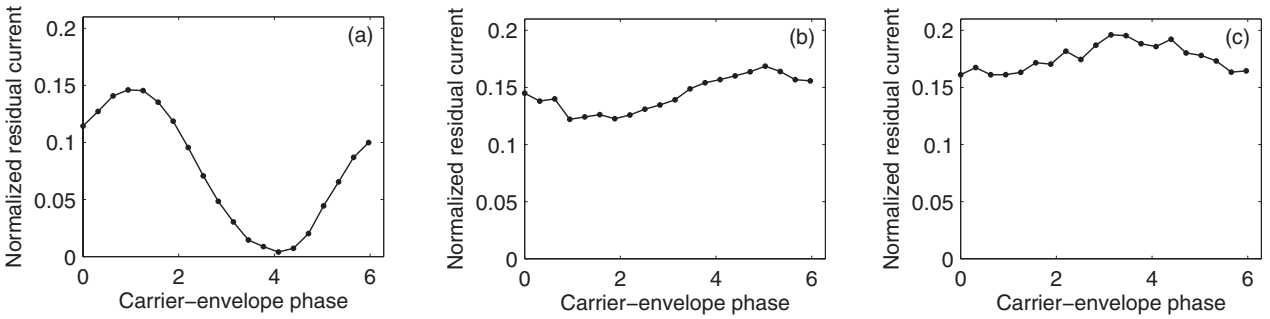

FIG. 1. CEP dependence of the normalized residual current for various pulse durations: (a) τ = three cycles, (b) τ = six cycles, and (c) τ = nine cycles. Laser peak intensity is 5×10^{15} W/cm^2; the molecular parameters are $D = 3.4a_0$ and $Z_1/Z_2 = 2$.

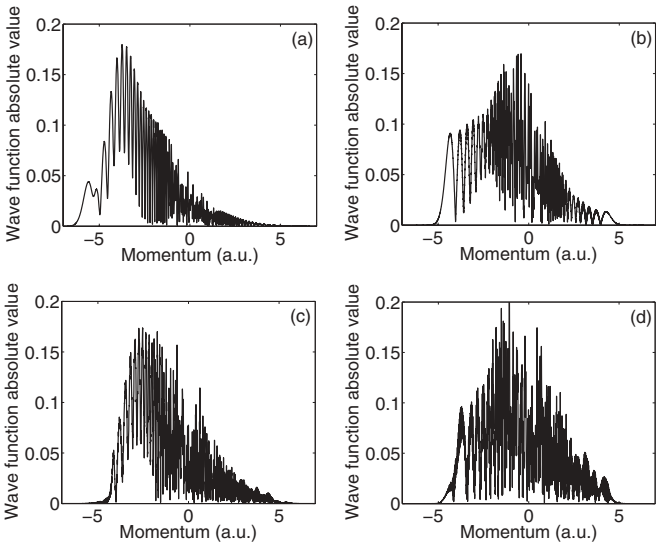

FIG. 3. Electron wave function in the momentum space after the passage of the three- (a) and nine- (c) cycle laser pulse in the case of an asymmetric molecule (charge ratio, 2:1). The results for a symmetric molecule with the same pulse durations are also shown, (b) and (d), respectively.

Figure 3 shows the electron wave packet in the momentum space after the passage of the laser pulse. Figures 3(a)–3(d) are plotted for parameters corresponding to maxima in Figs. 2(a)–2(d), respectively. This figure shows, in particular, how the average momentum of the electron changes with the parameters Z_1/Z_2 and τ. It is clearly seen that in the case of an asymmetric molecule, the wave packet is stronger shifted from zero. With increasing duration of the laser pulse, the momentum distribution for a symmetric molecule gets more symmetrized with respect to zero, while in the case of an asymmetric molecule it remains skew.

Figure 4 shows the dependence of the normalized residual current on the laser pulse full width at half maximum (FWHM $\approx 0.364\tau$) and the degree of the charge asymmetry; the other parameters are optimized. From this figure, it is clear that the asymmetry of the molecular potential at fixed other parameters always leads to higher efficiency of the directional current generation with respect to the symmetric case; the greater the asymmetry of the potential is, the more significant the gain in efficiency is. While for a symmetric system the residual current goes to zero with increasing pulse duration, for an asymmetric molecule it demonstrates a saturation behavior.

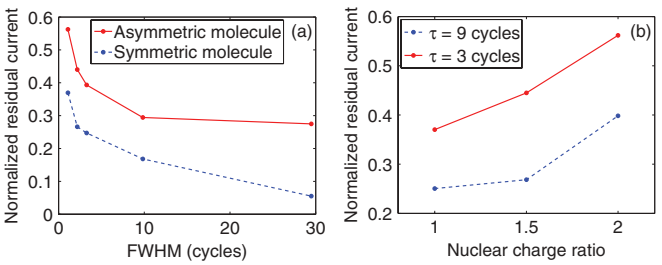

FIG. 4. (Color online) Normalized residual current as a function of (a) laser pulse duration and (b) degree of the charge asymmetry; the other parameters of the laser pulse and quantum system are optimized.

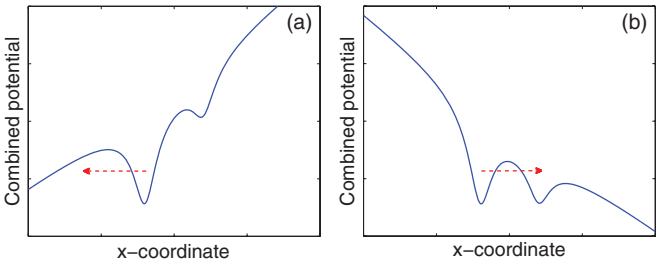

FIG. 5. (Color online) Combined Coulomb and laser field potentials for the asymmetric molecule (charge ratio, Z_1:Z_2 = 2:1) at the time instants when the external field direction is (a) positive and (b) negative. The arrows point to the direction of the electron escape.

III. MECHANISM OF QUASI-dc CURRENT EXCITATION IN THE ASYMMETRIC POTENTIAL

Figure 5 shows the instantaneous combined Coulomb and laser field potentials $V(x) + E(t)x$ for the asymmetric molecule (charge ratio, Z_1:Z_2 = 2:1) at two time instants separated by a half-cycle of the laser field.

In the case of an asymmetric molecule, the electron ground-state wave function is localized mainly on the deeper potential well at the nucleus with larger charge (left well in Fig. 5). When the electric field has a positive direction, forcing the electron to move to the left [Fig. 5(a)], the situation is similar to the atomic case. In this case, in order to escape from the well, the electron should tunnel through a wide external barrier. In the adjacent half-cycle, the electric field is negative, causing the electron to move to the right [Fig. 5(b)]. In this case, the electron tunnels through the internal barrier, whose width is strongly dependent on the internuclear distance and can be much narrower than in the former (atomiclike) case. As a consequence, the electron ionization probability can differ dramatically between the adjacent half-cycles. The maximum difference is reached at internuclear distances for which enhanced ionization [20,21] is observed.

To simplify further discussion, let us consider a quasi-monochromatic laser pulse. Figure 6 shows one cycle of this pulse along with the arrows showing schematically the drift velocity of the electrons released at different times.

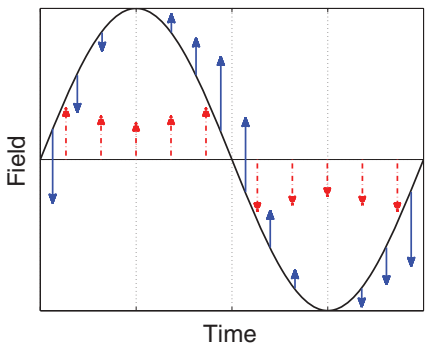

FIG. 6. (Color online) Schematic picture showing the drift velocity acquired by an electron by the end of the laser pulse, depending on the time of the electron release. The partial contributions due to the laser field and Coulomb potentials are depicted by blue solid and red dash-dotted arrows, respectively.

If the Coulomb potential of the parent ion ceased to act on the electron immediately after the ionization instant, the electron drift velocity would be constant (shown by blue solid arrows in Fig. 6) and determined only by the magnitude of the vector potential of the laser at the time of the electron release. In fact, after escaping from the well the electron continues to be influenced by the Coulomb potential, and this effect is quite strong while the electron is not removed to a sufficiently long distance from the ionic core. Thus, the influence of the Coulomb potential leads to an additional change in the electron drift velocity (red dash-dotted arrows in Fig. 6). If the symmetry of the ionization process is not broken, the Coulomb corrections to the drift velocity of the electrons released at times differing by a half cycle of the field are equal in magnitude and opposite in sign. As a result, the average electron drift velocity is equal to zero (as if the Coulomb potential did not act on a free electron). In contrast, in the case of an asymmetric potential discussed above, the electron tunneling in the half-cycles of the field, for which the electric field is positive [Fig. 5(a)], is much less probable than in adjacent half-cycles [Fig. 5(b)]. As a limiting case, the laser pulse can be thought of as acting on the electrons only during the half-cycles when the field is negative. In Fig. 6, this can be depicted by discarding arrows in the left half of the image. However, even with such an asymmetrical ionization, the assumption that there is no influence of the Coulomb potential on a free electron still leads to the result that the total current is zero, since the elementary currents caused by the electron release in the third and fourth quarters of the field cycle cancel each other (see blue arrows). In contrast, since during this time interval the Coulomb correction to the electron drift velocity (being nonzero) has the same sign (see red arrows), for the entire cycle of the field the electrons acquire a nonzero average drift velocity, which leads to the generation of a macroscopic residual current.

To test the above mechanism of the residual current excitation, we performed a series of calculations in a simple classical model. In this model, it is assumed that the electron escapes in only one direction [as in Fig. 5(b)], and after tunneling through the barrier its motion in the combined Coulomb and laser field potentials can be described classically. In addition, it is assumed that the processes of electron elastic rescattering can be neglected, that is, that the Coulomb correction to the drift velocity is accumulated solely in the first departure of an electron from the parent ion. The validity of this assumption was tested using three-dimensional quantum-mechanical calculations (see below). A comparison of the results of 1D classical and quantum-mechanical calculations in the models described above is presented in Fig. 7. As a quasi-monochromatic pulse, in these quantum-mechanical calculations we used the trapezoidal pulse with 3-cycle linear ramps and a 20-cycle interval of constant amplitude. When comparing the results, the residual current found in the classical calculations was multiplied by the total probability of molecular ionization found in the corresponding quantum-mechanical simulations.

The data presented in Fig. 7 show that the simple classical model proposed above provides quite a good description of the directional current generation in an asymmetric potential. The observed small discrepancies can be explained by two factors.

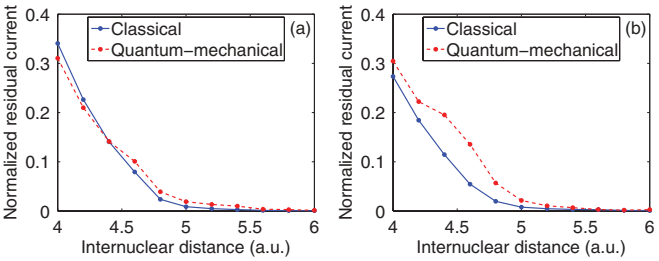

FIG. 7. (Color online) Dependence of the normalized residual current on the internuclear distance in the asymmetric molecule (charge ratio, Z_1:Z_2 = 2:1). Laser peak intensity is (a) 3.1×10^{15} W/cm^2 and (b) 3.8×10^{15} W/cm^2. The results of 1D classical (blue solid line) and quantum-mechanical (red dotted line) calculations are shown.

First, the classical model ignores the electron release during half-cycles in which the field is positive [Fig. 5(a)]. This leads to a slight overestimation of the residual current in the classical calculations. Second, for the half-cycles in which the field is negative [Fig. 5(b)], the classical model neglects the electron tunneling through the outermost barrier (only electrons whose removal from the right well is allowed classically are taken into account). This assumption results in some underestimation of the residual current in the classical calculations. For the range of parameters chosen to compare the data, the second factor has a greater impact on the final results, so the classical curves are usually below the quantum-mechanical ones.

IV. THREE-DIMENSIONAL NUMERICAL SIMULATION

For some cases, we have also carried out full three-dimensional (3D) quantum-mechanical numerical simulations. The results of 3D calculation of the normalized residual current as a function of the laser pulse duration are shown in Fig. 8. In the 3D calculations, all parameters were fixed equal to their optimal values found for the 1D case.

As can be seen from the comparison of Figs. 4(a) and 8, the 1D model generally overestimates the value of the residual current. On the other hand, since the parameters in the 3D case are not optimized, it can be expected that under optimal conditions the residual current is greater than that shown in Fig. 8. Anyway, both the 1D and 3D models in general give

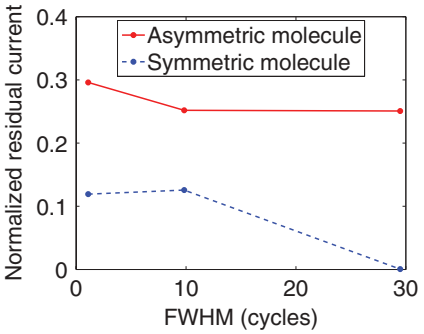

FIG. 8. (Color online) 3D results for the normalized residual current as a function of the laser pulse duration for asymmetric (Z_1:Z_2 = 2:1) and symmetric molecules.

the same behavior of the residual current, namely, its growth with decreasing laser pulse duration and increasing asymmetry of the molecular potential. We also note that in the more realistic 3D case, the asymmetry of the potential has even greater impact than in the 1D model. This can be explained by a different role played by the electron trajectories with multiple returns to the parent ion in the 1D and 3D cases. In the 1D case, with such returns, the electron acquires one by one the Coulomb corrections of opposite sign to the drift velocity, which may partly cancel each other. In the 3D case, because of the rapid wave-packet spreading, such compensation is much less significant.

Additionally, 3D calculations were performed for nonzero ellipticity of the laser field. The calculations revealed that the projection of a normalized residual current on the molecular axis is almost independent of the laser ellipticity, which confirms the assumption used in the classical model of no influence of the electron elastic rescattering from the parent ion on the magnitude of the directional current.

V. CONCLUSION AND DISCUSSION

In conclusion, we have shown that the magnitude of directional current and the efficiency of its excitation in tunneling ionization of asymmetric systems can be significantly enhanced compared to the case of symmetric systems. The results obtained in a simple classical model favor the Coulomb mechanism of high-efficiency generation of residual current, while, according to the 3D quantum-mechanical calculations, electron rescattering effects play a minor role. The results obtained also show that the optimization of directional current generation by reducing the duration of the laser pulse and the optimization of directional current by the use of asymmetric molecules are not mutually exclusive. Both approaches can be used simultaneously and, as follows from Figs. 4 and 8, give a greater overall effect.

Strong dependence of the directional current on the orientation of the molecular axis and the internuclear distance makes it possible to control the process of generation of terahertz radiation by producing rotational, vibrational, or dissociative wave packets, similar to what was proposed earlier for high-order harmonic generation in molecular gases [22–25].

Note also that the measurement of the dependence of the terahertz signal on the delay time can be used to probe the vibration-rotational dynamics of heteronuclear molecules. Because of the relative simplicity of these measurements, this method can be quite competitive with the more complex method based on the use of the Coulomb explosion [17], as well as with the methods exploiting the dependence of the high harmonic yield on the instantaneous structure [26,27] and orientation [28] of molecules.

The effect discussed in this paper, i.e., the Coulomb potential-induced residual current generated by ionization of heteronuclear molecules in a quasi-monochromatic field, has essentially a nature similar to that of the well-known effect of the Coulomb potential of the parent ion on the escaping electron giving rise to unusual electron angular and energy distributions in atomic above-threshold ionization (see, e.g., Refs. [29,30]). In this regard, we note also the very recent observation of the Coulomb asymmetry of electron angular distributions in strong-field multielectron ionization of diatomic molecules [31]. The nature of the observed asymmetry is very similar to what is discussed in this paper.

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