Shape of the cross section for electron ionization of laser-excited atoms

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Nixon and Murray [K. L. Nixon and A. J. Murray, Phys. Rev. Lett. **112**, 023202 (2014)] have measured electron ionization of the 3 $^1P_1$ state of ^{24}Mg. which was excited from the ground state by linearly polarized laser radiation. The direction of laser polarization was parallel to the scattering plane and based on their measurements, they proposed a functional form for the differential cross section as a function of the angle that the polarization direction makes with the incident electron beam. We show that this proposed functional form is exact and express the parameters of this form in terms of the T-matrix elements for ionization calculated in the scattering frame. We also show that this form occurs for radiation linearly polarized in an arbitrary direction. Furthermore, we similarly treat the case of excitation by circularly polarized light where a more complex shape is obtained, though this reduces to the form for linearly polarized radiation for ionization in the coplanar geometry. Using the measured results from Nixon and Murray, we calculate the ratio of the magnitudes of the T-matrix elements as well as the difference in their phases.

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I. INTRODUCTION

Recently, Nixon and Murray [1] have published results of their measurement of the ionization of laser-excited Mg atoms in the 3 $^1P_1$ state by electron impact. They noted that the cross section had a characteristic shape depending on the direction of the linear polarization vector of the laser radiation used to excite the atom and fitted their results to a parametrization of this shape. It is the purpose of this paper to show that the form of the cross section they used is correct and is a function only of the polarization direction and does not depend on the details of the ionization process itself. We also present more general results of both linearly and circularly polarized light in any direction.

II. LINEAR POLARIZATION

A. Coplanar geometry

We will assume that the orbital and spin quantum numbers L and S are separately conserved and the initial state of the atom has quantum numbers $L = S = 0$, although the analysis of this paper can be applied in more general cases. After excitation by the laser beam, the atom will still have $S = 0$, but with opposite parity and $L = 1$ for a nontrivial case [2].

The cross section for ionization is proportional to the square of the absolute value of the transition matrix (see [3], Eq. 14.157), which involves details of the ionization process. It can be written in postform as

$$T = \langle \Psi | V | \Phi_{LM} e^{i\boldsymbol{k}\cdot\boldsymbol{r}} \rangle, \tag{1}$$

where Ψ is the wave function of the total system in the final channel, Φ_{LM} is the wave function of the excited atom before ionization, V is the interaction potential between the incident electron and the atom, and the exponential term represents the incident electron with momentum vector $\boldsymbol{k}$. This is a complicated quantity to evaluate accurately, but we do not need to know its value for the present analysis. For a given ionization process with fixed energies and angles for the incident and outgoing electrons, it is sufficient to indicate that the transition matrix will depend on the magnetic quantum number M of the excited atom that is ionized. We will denote the matrix element by T_M.

In the experiment described in [1], the momentum vectors of the incident and two outgoing electrons all lie in a single plane, the scattering plane. This arrangement is referred to as coplanar geometry. We take the usual scattering coordinate system F, where the axis of quantization (the z axis) is along the direction of the incident electron and the y axis is perpendicular to the scattering plane. Then the x axis lies in the scattering plane. The laser radiation was linearly polarized and propagated perpendicular to the scattering plane in the direction of the y axis. If we take a coordinate system F' with the z' axis along the direction of the laser polarization and the y$'$ axis in the same direction as the y axis of the system S then the x' axis also lies in the scattering plane. If the angle between the z axis and z' axis is ε, then a rotation about the y axis through an angle $-\varepsilon$ (using the conventions in [1]) brings the z axis into coincidence with the z' axis (see Fig. 1).

Nixon and Murray [1] found that when only the angle ε of the direction of the polarization vector was varied and the energy and momenta of the free electrons were fixed, the ionization differential cross sections could be fitted to the parametric form

$$Q \propto B\{1 + P\cos[2(\varepsilon - \kappa)]\}, \tag{2}$$

where Q is the measured cross section and B, P, and κ are parameters that were determined from the measurements. The general shape of this function is shown in Fig. 2. Note that this function possesses reflection symmetry about both lines $\varepsilon = \kappa$ and $\kappa + \pi/2$.

Since the laser radiation is linearly polarized along the z' axis the excited state Φ'_{LM} will have quantum numbers $L = 1$, $S = 0$, and $M = 0$ in the F' coordinate system [2]. Following [4], we can transform this wave function to the F coordinate system via the relation

$$\Phi'_{10} = \sum_{M'=-1}^{1} \Phi_{1M'} \mathcal{D}^1_{M'0}(0,-\varepsilon,0) = -\frac{\sin\varepsilon}{\sqrt{2}}\Phi_{1-1} + \cos\varepsilon\,\Phi_{10} + \frac{\sin\varepsilon}{\sqrt{2}}\Phi_{11}, \tag{3}$$

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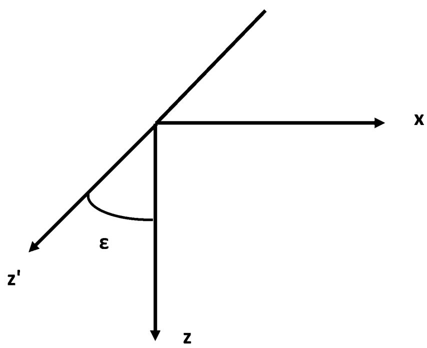

FIG. 1. Coordinate system for the experiments in [1]. The electrons were incident along the z axis and the outgoing electrons lie in the xz plane. The z' axis lies along the direction of polarization of the laser radiation used to excite the target atom before ionization. The y axis is perpendicular to the plane of the diagram and coincides with the direction of propagation of the laser beam. The angle ε is the angle between the z and z' axes and a rotation about the y axis through an angle $-\varepsilon$ brings the z axis into coincidence with the z' axis.

where $D^L_{M'M}(\alpha,\beta,\gamma)$ are the rotation matrix elements depending on the Euler angles (α,β,γ) required to rotate F into F'. Since in the present case all that is required is a single rotation to make the two coordinate systems coincide, $\alpha = \gamma = 0$. We have taken the specific values for these rotational matrix elements from Table I of [4].

Inserting (3) into (1), we find that the T matrix for the ionization of the atom excited by laser radiation with polarization axis in the scattering plane at an angle ε to the incident electron is given by

$$T = \cos\varepsilon T_0 + \frac{\sin\varepsilon}{\sqrt{2}}(T_1 - T_{-1}), \tag{4}$$

where T_M is the T-matrix element for ionization of the $3\,{}^1P_1$ state with magnetic quantum number M calculated in the F coordinate system. Since the cross section is proportional to the square of the absolute value of the T matrix (see [3], Eq. 15.24), we have

$$\begin{aligned}|T|^2 &= \cos^2\varepsilon|T_0|^2 + \frac{\sin^2\varepsilon}{2}|T_1 - T_{-1}|^2 \\ &\quad + \sqrt{2}\cos\varepsilon\sin\varepsilon\,\mathrm{Re}[T_0^*(T_1 - T_{-1})] \\ &= \left[\frac{|T_0|^2}{2} + \frac{|T_1 - T_{-1}|^2}{4}\right] + \left[\frac{|T_0|^2}{2} - \frac{|T_1 - T_{-1}|^2}{4}\right]\cos 2\varepsilon \\ &\quad + \frac{1}{\sqrt{2}}\mathrm{Re}[T_0^*(T_1 - T_{-1})]\sin 2\varepsilon,\end{aligned} \tag{5}$$

where Re indicates the real part of the bracketed quantity and the asterisk denotes the complex conjugate. Then the right-hand side of (5) can be put into the form (2) with

$$B = \frac{|T_0|^2}{2} + \frac{|T_1 - T_{-1}|^2}{4}, \tag{6}$$

$$P = \frac{1}{2}\left|T_0^2 + \frac{(T_1 - T_{-1})^2}{2}\right|/B, \tag{7}$$

$$\tan 2\kappa = 2\sqrt{2}\frac{\mathrm{Re}[T_0^*(T_1 - T_{-1})]}{2|T_0|^2 - |T_1 - T_{-1}|^2}. \tag{8}$$

The quadrant in which the angle 2κ lies is determined by the sign of the numerator and denominator in (8) separately. It is important to note that it is only necessary to calculate the T matrices for the ionization of each of the magnetic substates of the initial excited state once in the collision frame (F coordinate system) in order to determine the cross section for an arbitrary direction of the polarization of the exciting radiation in the scattering plane.

Since the momenta of the incident and emerging electrons are coplanar and the spins are unresolved, the experiment of Nixon and Murray [1] possesses mirror symmetry in the scattering plane. Thus there is the relationship $T_{-1} = -T_1$ between the T-matrix elements [5]. This simplifies the above parameters somewhat and reduces number of T-matrix elements that need to be calculated.

In Fig. 4 of [1] values for the parameters P and κ are plotted as a function of the angle of the ejected electron. The angle

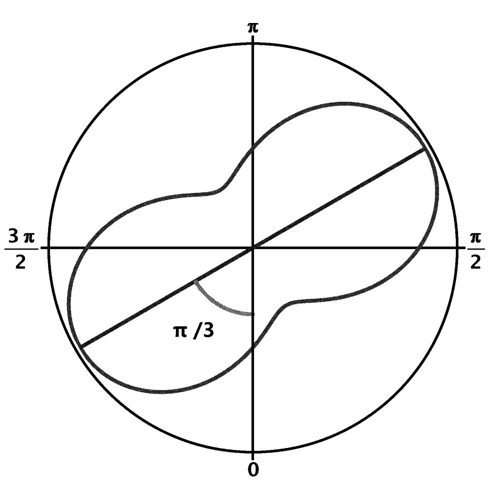

FIG. 2. Plot of the function in Eq. (2) with $B = 2$, $P = \frac{1}{2}$ and $\kappa = -60°$.

TABLE I. Two solutions for the ratio $R = |T_1|/|T_0|$ and the value of $\cos\Delta$ corresponding to the first solution, where Δ is the difference in phases of T_0 and T_1 given as a function of the angle θ of the ejected electron as calculated from Eqs. (7) and (8). The value of $\cos\Delta$ corresponding to the second solution is just the negative of the one shown. The values used for P and κ are shown in Fig. 4 of [1].

θ	R_1	R_2	$\cos\Delta$
40.0	0.32	1.55	0.00
45.0	0.36	1.40	0.10
50.0	0.45	1.10	0.41
55.0	0.60	0.83	0.37
57.5	0.46	1.08	–0.15
60.0	0.45	1.11	0.00
65.0	0.37	1.37	0.08
70.0	0.32	1.54	0.41
75.0	0.22	2.27	0.31
80.0	0.20	2.56	0.54

of the scattered electron was fixed at 30° and both outgoing electrons had energies of 20 eV. Since the ionization potential of the excited 3 $^1P_1$ state of the target atom is 3.30 eV, the incident electron had an energy of 43.30 eV. Using these values of P and κ in Eqs. (7) and (8) and the relationship due to mirror symmetry discussed above, we can calculate the ratio of the magnitude of the T-matrix elements $R = |T_1|/|T_0|$ and $\cos\Delta$, where Δ is the difference in phases of T_0 and T_1.

Let $S = \tan 2\kappa$ and $W = (\sqrt{1+S^2} - P)/(\sqrt{1+S^2} + P)$. Then there are two positive solutions for R given by $R_1 = \sqrt{W/2}$ and $R_2 = 1/\sqrt{2W}$. The value of $\cos\Delta$ corresponding to R_1 is given by $(1-W)S/2\sqrt{W}$, while the value corresponding to R_2 is the negative of this. Note that since $0 < P < 1$, the sign of $\cos\Delta$ is determined by the sign of S. The values of R and $\cos\Delta$ calculated from the data in Fig. 4 of [1] are given in Table I. The calculations ignore the error bars in the experimental results.

We note that when $\kappa = 45°$ the left-hand side of Eq. (8) is infinite, which implies that $R = 1/\sqrt{2}$. From Fig. 4 of [1] this occurs for a value of κ between 32° and 80°. In order for Δ to be continuous, the solution used for R must switch from R_1 to R_2 at this point or vice versa.

B. Noncoplanar geometry

We now consider the case where the laser radiation is linearly polarized in an arbitrary direction with respect to the scattering coordinate system F. Furthermore, the scattering plane referred to in the previous section can be defined by the two vectors representing the momentum of the incident and scattered electron with the direction of the ejected electron being arbitrary. We will again assume that LS coupling is valid and that the atomic state before excitation with the laser radiation is given by $L = 0$ but arbitrary S. Further, after excitation, we assume that the atom has $L = 1$ with opposite parity to the initial states and the same spin S. As before, since the exciting radiation is linearly polarized, the magnetic quantum number $M = 0$. If the direction of polarization is specified by the positive angles (ε,γ) then we can define the coordinate system F' as having the z' axis along this direction of polarization and the y' axis as perpendicular to the plane containing both the z and z' axes pointing in directions such that a rotation about this axis through an angle $-\varepsilon$ brings the z axis into coincidence with the z' axis (see Fig. 3).

The direction of the laser beam will be perpendicular to the z' axis but otherwise unspecified. In this case a further rotation about the z axis through an angle $-\gamma$ is necessary to make the coordinate system F coincide with F'. Following the analysis of the previous section, the transformation given in Eq. (3) will become

$$\Phi'_{10} = \sum_{M'=-1}^{1} \Phi_{1M'}\mathcal{D}^1_{M'0}(0,-\varepsilon,-\gamma) = -\frac{\sin\varepsilon}{\sqrt{2}}\Phi_{1-1} + \cos\varepsilon\,\Phi_{10} + \frac{\sin\varepsilon}{\sqrt{2}}\Phi_{11}. \quad (9)$$

Since the right-hand side of this equation is the same as in Eq. (3) the results of the previous section will remain valid and in particular the dependence of the cross section on ε will be given by Eq. (2) with no dependence on the angle γ. In general, the T-matrix elements will be different, especially if the momentum vector of the ejected electron does not lie in the scattering plane as defined above.

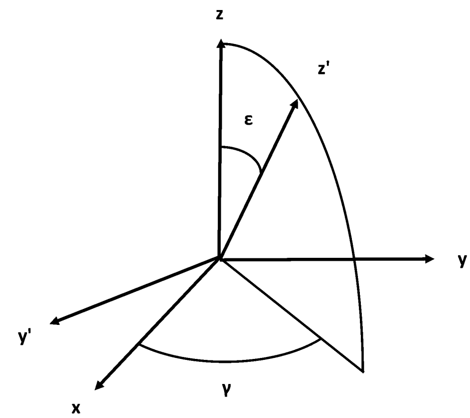

FIG. 3. Coordinate system in the case where the radiation is polarized in an arbitrary direction along the z' axis. As before, the electrons are incident along the z axis and the incident and scattered electron momenta lie in the xz plane while the direction of the ejected electron is arbitrary. This coordinate system also represents the case where the exciting radiation is circularly polarized. In this case, the z' axis represents the direction of propagation of the radiation.

III. CIRCULAR POLARIZATION

Using circularly polarized laser radiation in place of the linear polarization case dealt with above, it is possible to excite an atom to a magnetic substate with $M = \pm L$. In this case the axis of quantization is along the direction of the laser beam rather than perpendicular to it, as was the case for linear polarization. We will deal with the more general case presented in the previous section but with $L = 1$ and $M = \pm 1$. However, the general angles (ε,γ) now refer to the direction of the laser beam rather than the direction of the polarization. We define the coordinate system F' similar to the way we defined it previously with the difference that the z' axis is now along the direction of the laser beam. Thus the rotations necessary to make the coordinate systems F and F' coincide are the same. In this case the excited atomic state before ionization with $M = 1$ can be represented as

$$\Phi'_{11} = \sum_{M'=-1}^{1} \Phi_{1M'}\mathcal{D}^1_{M'1}(0,-\varepsilon,-\gamma) = e^{i\gamma}\left[\sin^2\left(\frac{\varepsilon}{2}\right)\Phi_{1-1} - \frac{\sin\varepsilon}{\sqrt{2}}\Phi_{10} + \cos^2\left(\frac{\varepsilon}{2}\right)\Phi_{11}\right], \quad (10)$$

which gives a T matrix calculated in the F coordinate frame

$$T = e^{i\gamma}\left[\sin^2\left(\frac{\varepsilon}{2}\right)T_{-1} - \frac{\sin\varepsilon}{\sqrt{2}}T_0 + \cos^2\left(\frac{\varepsilon}{2}\right)T_1\right], \quad (11)$$

where again T_M refers to the T matrix calculated in the F coordinate system for the excitation of an atom with angular momentum quantum numbers LM in this system. The squared modulus of the T matrix that is proportional to the differential cross section can be put in the form

$$|T|^2 = A + U\cos(\varepsilon - \delta) + V\cos[2(\varepsilon - \kappa)], \tag{12}$$

where

$$A = \tfrac{1}{4}|T_0|^2 + \tfrac{1}{4}|T_1 + T_{-1}|^2 + \tfrac{1}{8}|T_1 - T_{-1}|^2, \tag{13}$$

$$U^2 = \tfrac{1}{4}[|T_1|^2 - |T_{-1}|^2]^2 + \tfrac{1}{2}\{\mathrm{Re}[T_0^*(T_1 + T_{-1})]\}^2, \tag{14}$$

$$V^2 = \left(\frac{|T_1 - T_{-1}|^2}{8} - \frac{|T_0|^2}{4}\right)^2 + \frac{1}{8}\{\mathrm{Re}[T_0^*(T_1 - T_{-1})]\}^2, \tag{15}$$

$$\tan\delta = \frac{-\sqrt{2}\mathrm{Re}[T_0^*(T_1 + T_{-1})]}{|T_1|^2 - |T_{-1}|^2}, \tag{16}$$

$$\tan 2\kappa = \frac{-2\sqrt{2}\mathrm{Re}[T_0^*(T_1 - T_{-1})]}{|T_1 - T_{-1}|^2 - 2|T_0|^2}. \tag{17}$$

As indicated in (16) and (17), the negative sign is associated with the numerator, which is required in order to determine in which quadrant the angles lie. A plot of (12) is rather complex in general and possesses none of the symmetries noted in the case of linear polarization. However, in the case of experiments in the coplanar geometry where $T_{-1} = -T_1$, $U = 0$, so (12) now has the same form as (2) with its associated symmetries.

The case where the magnetic sublevel $M = -1$ is excited by circularly polarized laser radiation can be most easily dealt with by replacing ε by $\varepsilon + \pi$. Thus the second term of (12) changes sign, but the equation is otherwise unchanged.

IV. CONCLUSION

In all cases of the ionization of laser excited atoms using LS coupling with $L = 1$ where the laser radiation is linearly polarized or the laser radiation is circularly polarized and the scattering system possesses mirror symmetry (coplanar geometry), the general shape of the cross section is given by Eq. (2). Only in the case of excitation by circularly polarized light where the ionization is noncoplanar will the shape of the cross section be given by the more general form (12).

An important consequence of these results is that the T-matrix elements for ionization only need to be calculated in the scattering frame whatever the direction or polarization of the exciting radiation. Thus this problem is computationally no more complex than the case of ionization of excited atoms where the quantization axis is along the direction of the incident electron. Situations where the initial excited atom has other values of orbital angular momentum L or obeys other coupling schemes can be dealt with in a similar manner. The characteristic shape of the cross section as a function of the direction of polarization of the laser radiation is independent of the method used to calculate the T-matrix elements and furthermore provides a check on any experimental measurements.

ACKNOWLEDGMENTS

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