

RAPID COMMUNICATION

Angular anisotropy parameters and recoil ion momentum distribution in two-photon double ionization of helium.

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Abstract. We present convergent close-coupling (CCC) calculations of the angular anisotropy parameters β_2, β_4 and the recoil ion momentum distribution $d\sigma/d\mathbf{p}$ in two-photon double ionization (TPDI) of helium. In a stark contrast to single-photon double ionization (SPDI) where the β_2 parameter varies widely changing the angular distribution from isotropic to nearly dipole for slow and fast photoelectrons, respectively, the β parameters for TPDI show very little change. The angular distribution of the recoil ion is fairly isotropic in TPDI as opposed to strong alignment with polarization of light in SPDI.

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There has been a considerable amount of research done recently on two-photon double ionization (TPDI) of He. This work was strongly driven by rapid advancement in experimental techniques. The Free electron LASer at Hamburg (FLASH) started delivering vacuum ultraviolet photons at intensities sufficient for observing various multi-photon phenomena. Two-photon single ionization of He at $\omega = 13$ eV was investigated by Laarmann *et al* (2005). Wabnitz *et al* (2005) studied multiple ionization of Ar and Xe atoms at the same photon energy. Moshhammer *et al* (2007) reported few-photon multiple ionization of Ne and Ar at $\omega = 38.8$ eV. Very recently, the same group obtained the first differential data on the sum-momentum distribution of TPDI of He at $\omega = 45$ eV (Moshhammer 2007). Using an alternative high harmonic generation technique, Nabekawa *et al* (2005) observed the production of doubly charged He ions by two-photon absorption using $\omega = 42$ eV radiation. Very recently, Benis *et al* (2006) observed two-photon double ionization of Ar and Kr atoms by a superposition of harmonics.

On the theoretical side, a wide range of methods was applied to the TPDI problem during the past decade. There have been several reported calculations of the total integrated cross-section of TPDI of He at various photon energies (Pindzola and Robicheaux 1998, Nikolopoulos and Lambropoulos 2001, Parker *et al* 2001, Mercouris *et al* 2001, Colgan and Pindzola 2002, Feng and van der Hart 2003, Piraux *et al* 2003, Hu *et al* 2005). Although numerical values of the cross-sections varied depending on the theoretical model and assumed characteristics of the laser field, there appeared some consensus between several calculations (see Hu *et al* (2005) for detail). This consensus, however, was challenged recently as can be seen from the latest reports (Foumouo *et al* 2006, Ivanov and Kheifets 2006, Nikolopoulos and Lambropoulos 2007).

In contrast, the data on angular correlation pattern in two-electron continuum are more conciliatory. There is at least some agreement between the fully resolved triply differential cross-sections (TDCS) produced by non-perturbative (Colgan and Pindzola 2002, Hu *et al* 2005) and perturbative (Kheifets and Ivanov 2006, Istomin *et al* 2006) calculations. Less detailed double differential cross-section (DDCS), integrated over the escape angle of one of the photoelectrons, was not explored so far. The only report of DDCS by Barna *et al* (2006) is concerned with the sequential regime of TPDI which shows much less sensitivity to many-electron correlations.

In the meantime, DDCS is much more appealing for experimental studies because of a larger volume of phase space being probed and significantly improved statistics. It can bring a wealth of information as was demonstrated by Knapp *et al* (2002) who were able to isolate various mechanisms of single-photon double ionization (SPDI) of He. Similar angular distributions differential with respect to the sum or difference momenta of the photoelectron pair can also be readily obtained (Dörner *et al* 1996, Bräuning *et al* 1997a, Knapp 2002). They proved to be useful in pinpointing propensity rules which govern the two-electron escape in the Jacobian coordinates (Walter *et al* 2000).

In this Rapid Communication, we bridge the gap between the total integrated and fully differential cross-sections of TPDI of He and present the calculations of the DDCS

in conventional and Jacobian coordinates. The latter calculation allows us to obtain the recoil ion momentum distribution which can be compared directly with a recent FLASH experiment (Moshhammer 2007). We use essentially the same dynamical model as was applied in our previous works on TPDI of He (Kheifets and Ivanov 2006, Kheifets *et al* 2007). In this model, the atom-field interaction is restricted to the lowest second-order perturbation theory which is further simplified by the closure approximation. In the meantime, the electron-electron integration is treated non-perturbatively and included in full. Although the closure approximation restricts the ability of this model to provide accurate absolute cross-sections, the angular correlation pattern obtained in such a calculation is remarkably similar to non-perturbative time-dependent close-coupling calculations (Colgan and Pindzola 2002, Hu *et al* 2005). We thus believe that this model should be adequate for DDCS calculations.

We start our derivation from the TPDI TDCS differential with respect to the photoelectron angles and energy:

$$\frac{d^3\sigma_M}{d\Omega_1 d\Omega_2 dE_2} \propto \left| \sum_{L=0,2} \sum_{\substack{l_1 m_1 \\ l_2 m_2}} C_{l_1 m_1}^{LM} Y_{l_1 m_1}(\hat{\mathbf{k}}_1) Y_{l_2 m_2}(\hat{\mathbf{k}}_2) (-i)^{l_1+l_2} e^{i(\delta_{l_1}+\delta_{l_2})} D_{l_1 l_2}^L(E_1, E_2) \right|^2 \quad (1)$$

Here the index M refers to the polarization state of light, $M = 0$ and 2 correspond to linear and circular polarization, respectively. In the latter case, only the quadrupole channel $L = 2$ contributes to TDCS. The radial matrix elements $D_{l_1 l_2}^L(E_1, E_2)$ are obtained in the simplified second-order perturbation theory as described in earlier work (Kheifets and Ivanov 2006). By integrating Equation (1) over $d\Omega_1$, we get DDCS which can be presented in the form

$$\frac{d^2\sigma}{d\Omega_2 dE_2} \propto \sum_{J=0,2,4} \beta_J P_J(\cos \theta_2) \quad (2)$$

where the β coefficients are given by the following expression:

$$\begin{aligned} \beta_J = & \hat{J}^2 \sum_{l_1} \sum_{\substack{l_2 l_2' \\ LL'}} \hat{L} \hat{L}' \hat{l}_2 \hat{l}_2' (-1)^{l_1+J} (-i)^{l_2-l_2'} e^{i(\delta_{l_2}-\delta_{l_2'})} \\ & \times \begin{pmatrix} L & J & L' \\ M & 0 & -M \end{pmatrix} \begin{pmatrix} l_2 & J & l_2' \\ 0 & 0 & 0 \end{pmatrix} \left\{ \begin{matrix} L & J & L' \\ l_2' & l_1 & l_2 \end{matrix} \right\} \text{Re} \left\{ D_{l_1 l_2}^L (D_{l_1 l_2'}^{L'})^* \right\} \end{aligned} \quad (3)$$

Here we introduced the hat symbol $\hat{L} = (2L+1)^{1/2}$ and dropped the explicit energy dependence of the matrix elements $D_{l_1 l_2}^L(E_1, E_2)$ for brevity. In deriving Equation (2) and (3), we coupled the angular momenta of the photoelectron pair L, L' into J using the graphical angular momentum projection summation technique (Varshalovich 1988). Equation (3) coincides with an analogous expression for β -parameters of two-photon single ionization (Liu *et al* 1992, Gribakin *et al* 1999) in which the role of l_1 is assumed by the angular momentum of the bound target electron. In the following, we divide the β -parameters by the single-differential, with respect of energy E_2 , cross-section such that $\beta_0 = 1$ and write the DDCS as

$$\frac{d^2\sigma}{d\Omega_2 dE_2} = \frac{d\sigma}{dE_2} \frac{1}{4\pi} [1 + \beta_2 P_2(\cos \theta_2) + \beta_4 P_4(\cos \theta_2)] \quad (4)$$

In comparison, the DDCCS of SPDI contains only the β_2 term which is given by the same Equation (3) with $L = L' = 1$.

To derive DDCCS in the Jacobian coordinates, we use the method proposed by Pont and Shakeshaft (1996) for SPDI. We start from the TPDI amplitude in the form suggested by Istomin *et al* (2006):

$$F(\mathbf{k}_1, \mathbf{k}_2) = f_1(\hat{\mathbf{k}}_1 \cdot \mathbf{e})^2 + f_2(\hat{\mathbf{k}}_2 \cdot \mathbf{e})^2 + f_s(\hat{\mathbf{k}}_1 \cdot \mathbf{e})(\hat{\mathbf{k}}_2 \cdot \mathbf{e}) + f_0(\mathbf{e} \cdot \mathbf{e}) \quad (5)$$

Here $\hat{\mathbf{k}}_i = \mathbf{k}_i/k_i$, $i = 1, 2$ are the unit vectors directed along the photoelectron momenta. The amplitudes f_i , $i = 0, 1, 2, s$ depend on the essential dynamic variables E_1 , E_2 and $\cos \theta_{12} = \mathbf{k}_1 \cdot \mathbf{k}_2/(k_1 k_2)$. We rewrite Equation (5) relative to the Jacobian momenta $\mathbf{p} = \mathbf{k}_1 + \mathbf{k}_2$, $\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$:

$$F(\mathbf{p}, \mathbf{k}) = f_p(\mathbf{p} \cdot \mathbf{e})^2 + f_k(\mathbf{k} \cdot \mathbf{e})^2 + f_{kp}(\mathbf{k} \cdot \mathbf{e})(\mathbf{p} \cdot \mathbf{e}) + f_0(\mathbf{e} \cdot \mathbf{e}) \quad (6)$$

where

$$\begin{cases} f_p &= \frac{1}{4} \left(\frac{f_1}{k_1^2} + \frac{f_2}{k_2^2} + \frac{f_s}{k_1 k_2} \right) \approx \frac{1}{4} \left[f \left(\frac{1}{k_1^2} + \frac{1}{k_2^2} \right) + \frac{f_s}{k_1 k_2} \right] \\ f_k &= \frac{1}{4} \left(\frac{f_1}{k_1^2} + \frac{f_2}{k_2^2} - \frac{f_s}{k_1 k_2} \right) \approx \frac{1}{4} \left[f \left(\frac{1}{k_1^2} + \frac{1}{k_2^2} \right) - \frac{f_s}{k_1 k_2} \right] \\ f_{kp} &= \frac{1}{2} \left(\frac{f_1}{k_1^2} - \frac{f_2}{k_2^2} \right) \approx \frac{1}{2} f \left(\frac{1}{k_1^2} - \frac{1}{k_2^2} \right) \end{cases}$$

Here we used a weak asymmetry approximation $f_1 \approx f_2 \equiv f$ thus neglecting the energy sharing dependence of the amplitudes.

Directing the (linear) polarization axis along z and noting that $\mathbf{p} \cdot \mathbf{e} = p \cos \theta_p$ and $\mathbf{k} \cdot \mathbf{e} = k \cos \theta_k$ we write squared amplitude (6) as

$$\begin{aligned} |F(\mathbf{p}, \mathbf{k})|^2 &= p^4 |f_p|^2 \cos^4 \theta_p + k^4 |f_k|^2 \cos^4 \theta_k + p^2 k^2 |f_{kp}|^2 \cos^2 \theta_p \cos^2 \theta_k + |f_0|^2 \\ &\quad + 2p^2 k^2 \operatorname{Re}\{f_p f_k^*\} \cos^2 \theta_p \cos^2 \theta_k + 2pk \operatorname{Re}\{f_{kp} f_0^*\} \cos \theta_p \cos \theta_k \\ &\quad + 2p^3 k \operatorname{Re}\{f_p f_{kp}^*\} \cos^3 \theta_p \cos \theta_k + 2pk^3 \operatorname{Re}\{f_k f_{kp}^*\} \cos \theta_p \cos^3 \theta_k \\ &\quad + 2p^2 \operatorname{Re}\{f_p f_0^*\} \cos^2 \theta_p + 2k^2 \operatorname{Re}\{f_k f_0^*\} \cos^2 \theta_k \end{aligned} \quad (7)$$

Next step is to expand the amplitude factors into the Legendre polynomial series with respect to the cosine of the mutual angle $\cos \theta_{kp} = (\mathbf{k} \cdot \mathbf{p})/kp$ as prescribed by Pont and Shakeshaft (1996). Using this expansion and the Legendre polynomials integral

$$\int d\Omega_k P_l(\hat{\mathbf{k}} \cdot \mathbf{e}) P_{l'}(\hat{\mathbf{k}} \cdot \hat{\mathbf{p}}) = \frac{4\pi}{2l+1} P_l(\hat{\mathbf{p}} \cdot \mathbf{e}) \delta_{ll'}$$

we can perform the spherical integration

$$\frac{1}{4\pi} \int d\Omega_k |F(\mathbf{p}, \mathbf{k})|^2 = A_0 + A_2 P_2(\cos \theta_p) + A_4 P_4(\cos \theta_p) \quad (8)$$

where the coefficients A_0 , A_2 and A_4 are expressed via the coefficients in the Legendre polynomials expansion of quadratic $|f_i|^2$, $i = p, k, kp, 0$ and bi-linear $\operatorname{Re}\{f_i f_j^*\}$ forms. The same technique can be applied directly to Equation (5) thus giving an alternative method of obtaining the β -coefficients in Equation (4). We used it as a check of the accuracy of our computations.

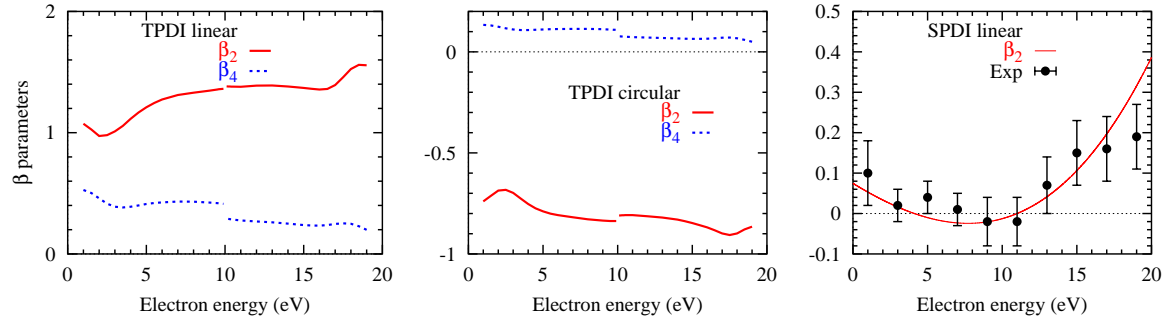


Figure 1. Angular anisotropy β parameters for two-photon (left and center) and single-photon (right) double ionization of He at the excess energy of 20 eV. Experimental data for SPDI are from Bräuning *et al* (1997b). The TPDI calculations are performed for linear and circular polarization of light.

The DDCS is the spherical integral (8) multiplied by an extra kinematical factor kp (Pont and Shakeshaft 1996):

$$\frac{d^2\sigma}{d\Omega_p dE_p} \propto kp \frac{1}{4\pi} \int d\Omega_k |F(\mathbf{p}, \mathbf{k})|^2 = \frac{d\sigma}{dE_p} \frac{1}{4\pi} [1 + \beta_2 P_2(\cos \theta_p) + \beta_4 P_4(\cos \theta_p)] \quad (9)$$

where

$$\frac{1}{4\pi} \frac{d\sigma}{dE_p} \propto kp A_0, \quad \beta_2 = \frac{A_2}{A_0}, \quad \beta_4 = \frac{A_4}{A_0}$$

We note that $d\sigma/dE_p = 0$ for $p = 0$ even though $A_0 \neq 0$ as the antiparallel escape resulting in $p = 0$ is not forbidden in TPDI as opposed to SPDI.

Now we turn to results of our calculations. On the left panel of Figure 1 we show the β parameters in Equation (4) for TPDI of He with linearly polarized light at the excess energy of 20 eV above the double ionization threshold which corresponds to the photon energy $\omega = 49.5$ eV. For comparison, on the right panel of the same figure, we show the β -parameter for SPDI at the same excess energy (the photon energy $\omega = 99$ eV). The experimental data for SPDI are from Bräuning *et al* (1997b).

In the CCC method, the electrons are explicitly distinguishable. The label 1 is attached to the slow electron which is described as a positive energy pseudostate whereas the label 2 corresponds to the fast electron which is considered as a Coulomb wave. Thus the expressions (3) and (4) define the angular distribution of the fast photoelectron $E_2 > E/2$ where E is the excess energy above the double ionization threshold. To obtain the angular distribution of the slow photoelectron, we rely on the exchange symmetry of the matrix elements $D_{l_1 l_2}^L(E_1, E_2) = D_{l_2 l_1}^L(E_2, E_1)$. Since the slow and fast electrons are described differently, numerical values of the direct and exchange matrix elements might be somewhat different. This is reflected in a small gap of β parameter at $E/2$.

By comparing the left and right panels of Figure 1, we observe a significant qualitative difference between the β parameters for TPDI and SPDI. In the two-photon case, both β_2 and β_4 are large and positive. In contrast, in the single-photon case, the β_2 parameter varies widely from nearly zero for a very slow photoelectron to large positive values for a fast photoelectron. This tendency to large β_2 is exemplified at larger excess

energies. For instance, at 100 eV excess energy, $\beta_2 \approx 1.4$ for the 99 eV photoelectron which demonstrates a strongly dipolar angular distribution (Knapp 2002). At larger still excess energy of 450 eV, $\beta_2 \approx 2$ at the top end of the excess energy scale which corresponds to the complete alignment of the fast photoelectron with the polarization axis of light (Knapp *et al* 2002). This energy dependence of β_2 can be interpreted as a gradual cross-over between two different regimes of SPDI. Nearly zero β_2 parameter of a slow photoelectron corresponds to a fully isotropic angular distribution which is a footprint of the shake-off mechanism. On the contrary, a large positive β_2 parameter for a fast photoelectron corresponds to a direct knock out when the whole of the photon energy and angular momentum is absorbed by the fast photoelectron.

This can be seen from the explicit expression of the β_2 parameter for SPDI which can be obtained from Equation (3) by setting $L = L' = 1$. When the shake-off is the main mechanism of the SPDI, the slow electron emerges predominantly in the s state. One therefore can truncate the sum in (3) by a single term $l_2 = l'_2 = 0$ which leads immediately to $\beta_2 = 0$. To find the β_2 parameter for a fast photoelectron, one can employ the exchange symmetry $D_{l_1 l_2}(E_1, E_2) = D_{l_2 l_1}(E_2, E_1)$ to establish the only surviving term corresponding to $l_1 = 0, l_2 = 1$ and $\beta = 2$.

In TPDI, because the target atom interacts with the field twice, the shake-off electron can be ejected in a p wave after firstly being promoted from the ground $1s$ state to an excited np state. In this scenario, the main contribution to the angular distribution of the slow electron comes from the $l_1 = l_2 = 1$ term which leads to $\beta_2 = 1$ and $\beta_4 = 0$. This is indeed close to the calculated value of β_2 and explains much smaller values of β_4 . By way of the exchange symmetry, one can argue that the same angular terms would be dominant for the fast electron which explains a rather weak energy dependence of β parameters. A non-zero value of β_4 indicates that the slow electron can also emerge in a d state as a result of two repeated knock out processes. Most likely, as in the case of SPDI, a clear separation of the shake-off and knock-out ionization mechanisms would occur at much higher photon energies. However, at this energies, the sequential regime of TPDI would become dominant.

The case of circular polarization which is illustrated in the central panel of Figure 1 can be analyzed similarly. In this case, a slow shake-off electron emerging in a p -state should have $\beta_2 = -1$ which is close our calculation. Although $\beta_4 \neq 0$, it remains small. As in the case of linear polarization, both β parameters show little variation with energy.

Now we turn our discussion to the angular distribution of the recoil ion momentum \mathbf{K} which is defined by the angular asymmetry parameters for the sum momentum $\mathbf{p} = \mathbf{k}_1 + \mathbf{k}_2 = -\mathbf{K}$. On the left panel of Figure 2 we plot the angular asymmetry β parameters and the energy distribution $d\sigma/dE_p$ corresponding to the sum momentum \mathbf{p} for the TPDI of He at the excess energy of 20 eV. For comparison, on the right panel of Figure 2, we plot the analogous parameters for the SPDI at the same excess energy. Again, as in Figure 1, there is a qualitative difference between the TPDI and SPDI cases. In the two-photon case, the β parameters change from nearly zero to large positive values as p varies from 0 to p_{\max} . This is to be compared with the large and

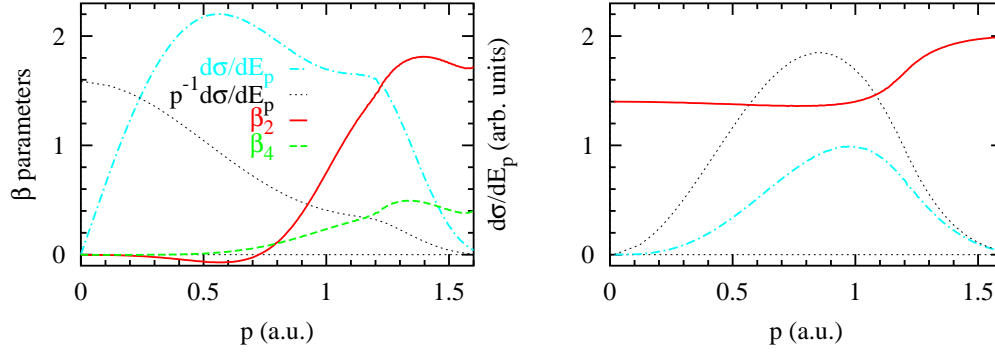


Figure 2. Angular anisotropy β parameters and energy distribution $d\sigma/dE_p$ of the recoil ion for single-photon (right) and two-photon (left) double ionization of He at the excess energy of 20 eV.

positive β_2 parameter which varies very little in SPDI. The energy distribution $d\sigma/dE_p$ is zero both for $p = 0$ and $p = p_{\max}$ ($k = 0$) due to the kinematic factor kp . However, the function $p^{-1}d\sigma/dE_p$ is large at $p = 0$ and decays monotonously towards p_{\max} in TPDI but has a broad maximum at about $\simeq p_{\max}/2$ in SPDI.

This qualitatively different behavior of β parameters can be explained in the following way. Inspection of Equation (7) shows that the terms containing various powers of $\cos\theta_p$ and thus contributing to A_2 and A_4 coefficients of (8) are all vanishing when $p \rightarrow 0$. In the meantime, A_0 remains finite as f_k is non-vanishing in this limit. This results in zero values of β_2 and β_4 and a finite value of $p^{-1}d\sigma/dE_p$ at $p = 0$. In comparison, the SPDI amplitude has the form $F(\mathbf{p}, \mathbf{k}) = f_p(\mathbf{p} \cdot \mathbf{e}) + f_k(\mathbf{k} \cdot \mathbf{e})$ where $f_{p,k} = 0.5(f_1/k_1 \pm f_2/k_2)$. When $p \rightarrow 0$, f_k goes to zero as well and both the coefficients A_0 and A_2 become small. This results in a finite β_2 and vanishing $p^{-1}d\sigma/dE_p$ at $p = 0$. We note that this analysis reflects the tensorial structure of the SPDI and TPDI amplitudes and thus remains valid for any atom other than He.

This different behavior of double ionization amplitudes in single- and two-photon case has a profound implication when the recoil ion momentum distribution is measured experimentally. In a typical cold target recoil ion momentum spectroscopy (COLTRIMS) experiment, the following quantity is recorded (Knapp 2002):

$$\frac{d\sigma}{dp_x dp_z} = \int_{-\Delta p_y}^{\Delta p_y} dp_y \frac{1}{p} \frac{d\sigma}{dE_p d\Omega_p}$$

Here the 3D momentum is projected on the polarization plane by way of integration over the momentum component p_y in the direction of the photon propagation. The limits of integration Δp_y are chosen depending on the signal count rate. For SPDI, thus observed momentum distribution will peak at $\simeq p_{\max}/2$ where β_2 is large and positive resulting in a broad dipole structure aligned along the polarization axis of light as is indeed the case for various excess energies ranging from 1 to 100 eV (Dörner *et al* 1996, Bräuning *et al* 1997a, Knapp 2002). On the contrary, in the TPDI case, the intensity of the momentum

distribution is largest near the origin where the β parameters are close to zero. Hence, there will be very little anisotropy seen in the recoil ion momentum distribution except for large momenta where the experimental signal is fairly weak. A recent COLTRIMS study of TPDI of Ne at photon energy of 38.8 eV confirmed this prediction (Moshhammer *et al* 2007). Unpublished preliminary results on He at 45 eV photon energy seem to also fit into this pattern (Moshhammer 2007).

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