Observation of anisotropic angular distribution of ionic fragments in the dissociation of CO^{2+}

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Angular distributions of fragment ions ($C^+ + O^+$) produced in dissociative double photoionization of carbon monoxide have been studied in the photon energy region 39–100 eV using a photoion-photoion coincidence technique and a source of synchrotron radiation. The asymmetry parameter β ranges from 0.69 at 39 eV to -0.19 at 100 eV. The observations of anisotropy in the dissociative double photoionization from the valence orbitals of CO are discussed in relation to the symmetry considerations involved in the transition.

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I. INTRODUCTION

In recent years, considerable attention has been directed to double and multiple photoionization of gaseous molecules. However, in spite of the growing experimental and theoretical interest in this field, there is still much to be learned concerning the details of production and dissociation dynamics of doubly charged molecules. In this regard, it is important to elucidate the symmetry of the electronic states of doubly charged ions and that of two electrons ejected which may result in anisotropic angular distributions of ionic fragments (symmetry-characterized dissociation). The angular distributions of fragments produced in molecular photodissociation are closely related to the symmetry and dynamics of the processes involved [1,2]. For the $\Sigma \rightarrow \Sigma$ transitions, the molecules oriented parallel to the light polarization vector are selectively excited, whereas for the $\Sigma \rightarrow \Pi$ transitions the molecules oriented perpendicular to the polarization vector are preferentially excited in randomly oriented free molecules. In the case of K-shell excitation and ionization, the symmetry of the intermediate states is well defined and the asymmetry parameter obtained experimentally for ionic fragments is consistent with some of the theoretical predictions [3–17]. On the contrary, however, in the case of direct double ionization (not via Auger decay), the ion pair angular distributions were believed to be essentially isotropic until very recently. This is because of the high density of the m^{2+} electronic states and of two outgoing waves of photoelectrons.

In 1991, two papers reported anisotropic angular distributions of fragment ions in direct double photoionization from valence orbitals; weak anisotropy (the asymmetry parameter $\beta \approx 0.2$, $h\nu = 37.5$ eV) in the dissociation of HBr^{2+} [18] and medium anisotropy ($\beta \approx 0.5$) in the dissociation of OCS^{2+} [19]. In the present study, angular distributions of fragment ions ($C^+ + O^+$) produced in the dissociative double photoionization of CO are determined by analyzing photoion-photoion coincidence (PIPICO) spectra. The present results are discussed in relation to the symmetry considerations involved in the transition.

II. EXPERIMENT AND DATA ANALYSIS

PIPICO spectra were measured in the region 39–100 eV using a rotatable time-of-flight (TOF) mass spectrometer and a constant-deviation grazing incidence monochromator together with synchrotron radiation at the ultraviolet synchrotron orbital radiation (UVSOR) facility of the Institute for Molecular Science, Okazaki. The TOF mass spectrometer and the associated electronic apparatus used in the present experiment are schematically shown in Fig. 1, details of which have been described elsewhere [20,21]. The photoion signals detected by a multichannel plate were fed into both the start and stop inputs of a time-to-amplitude converter (TAC). An electric field of 2250 V/cm was applied across the ionization region, from which ions produced were extracted through the first exit aperture (6 mm in diameter). Ions were further accelerated in the acceleration region (3750 V/cm) and entered into the 60-cm-long drift tube through the second exit aperture (10 mm in diameter). Aluminum optical filters were used to eliminate higher-order radiation. The PIPICO spectra were measured at 55°, 0°, and 90° relative to the polarization vector of the

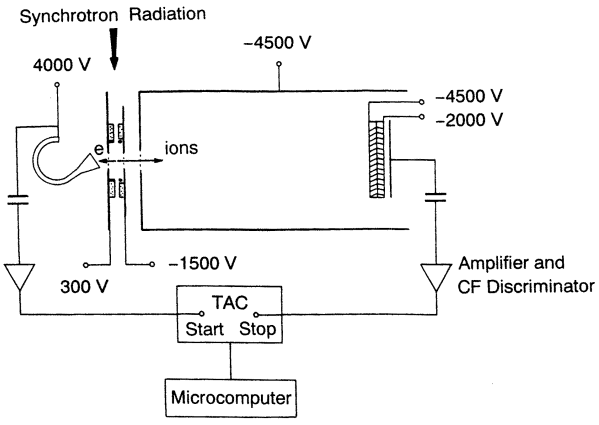

FIG. 1. Schematic diagram of the experimental apparatus. The ion detection signals were fed into both the start and stop inputs of a TAC for the measurements of PIPICO spectra. CF Discriminator denotes constant-fraction discriminator.

light in order to obtain the kinetic-energy release distributions (KERD's) and the angular distributions of the fragment ions.

In the framework of the dipole approximation for randomly oriented molecules with cylindrical symmetry in the gas phase, the differential partial cross section of the fragment ions for the partially polarized light is given [2,22] by

$$\frac{\partial\sigma_i}{\partial\Omega}=\frac{\sigma_i}{4\pi}\left[1+\frac{\beta}{4}(3p\cos2\theta+1)\right], \quad (1)$$

where β is the asymmetry parameter that characterizes the angular distribution, p is the degree of polarization, and θ is the angle of the spectrometer axis relative to the electric vector of the light. Depending on whether the transition moment is parallel ($\Delta\Lambda=0$) or perpendicular ($\Delta\Lambda=\pm1$) to the molecular axis (dissociation axis), the angular distribution is given by $\cos^2\theta$ ($\beta=2$) or $\sin^2\theta$ ($\beta=-1$), respectively, under the axial recoil conditions, i.e., the dissociation time is small enough compared to the rotational period ($\sim10^{-10}$ sec). The characteristic features of the simulated PIPICO spectral profiles obtained with the experimental conditions mentioned in Sec. II are illustrated in Fig. 2 for the $C^+ + O^+$ ion pair produced from CO^{2+} when the fragment ions have a single-valued kinetic energy of 2.6 eV. Figure 2 clearly indicates that the PIPICO spectral profile is a good measure of anisotropic angular distributions.

The spectral profiles of PIPICO peaks are determined by the kinetic-energy distribution and the angular distribution of the fragment ions, both relative to the spectrometer axis, and other experimental conditions such as the electric field across the ionization region and the size and degree of polarization of the photon beam. To obtain the angular distribution of fragment ions, the kinetic-energy release distribution was first determined by analyzing the PIPICO spectra measured at the so-called "pseudomagic angle," which is equal to about 55° under the assumption that $p=0.9$ [19]. The results will be reported separately [23]. The β parameter was obtained using the following three steps [19]: (1) Spectral profiles at $\theta=0°$ and 90° were calculated for a set of various kinetic energies of the fragment ions with β fixed arbitrarily; (2) the spectral profile of the PIPICO peak at angle θ was calculated based on the kinetic-energy distribution already determined and on the profiles calculated in step 1; and (3) treating β as a running parameter, its most probable value was determined as the one for which the sum of squares of the residuals between the observed and calculated profiles was minimized.

III. RESULTS AND DISCUSSION

Figure 3 shows two spectral profiles of the $C^+ + O^+$ PIPICO peaks produced from CO^{2+} at 39 eV and measured at $\theta=0°$ and 90°. The width of the PIPICO peaks results from the kinetic energy of the ion pair. The lighter ions (C^+) with a given momentum initially directed towards the flight tube will have longer flight-time differences than the C^+ ions initially directed away from the flight tube, thus resulting in the spread in flight-time differences observed for the ion pair. The profiles in Fig.

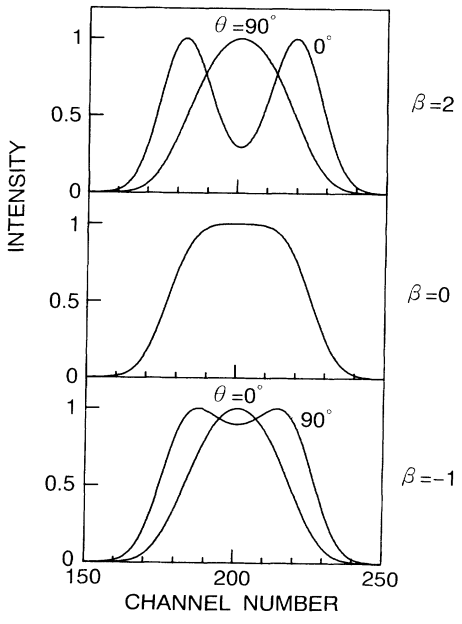

FIG. 2. Simulated PIPICO spectral profiles of the $C^+ + O^+$ ion pair with a single-valued kinetic energy of 2.6 eV at $\theta=0°$ and 90° for parallel ($\beta=2$), isotropic ($\beta=0$), and perpendicular ($\beta=-1$) transitions, which show characteristic features of anisotropic angular distributions. The central channel is assumed to be 200.

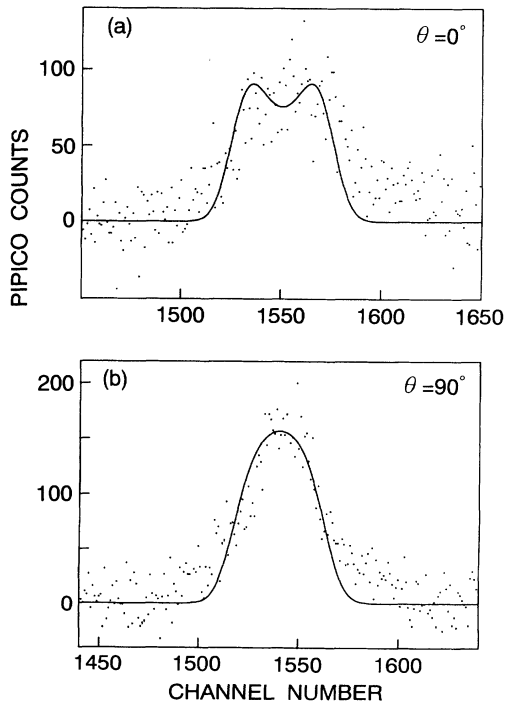

FIG. 3. Observed (dots) and simulated (solid lines) spectral profiles provide clear evidence of anisotropy in the $C^+ + O^+$ channel of CO^{2+} at 39 eV. (a) $\theta=0°$ ($\beta=0.64$) and (b) $\theta=90°$ ($\beta=0.73$). One KER component of 2.62 eV is assumed.

3 are both different from one another and from the profile shown in the middle of Fig. 2. This provides clear evidence of anisotropy of the ion pair. The best fit was attained for $\beta=0.64$ ($\theta=0°$) and 0.73 ($\theta=90°$). The observed β parameter obtained by analyzing the PIPICO peak shapes is shown in Fig. 4 as a function of excitation energy. Above 80 eV, the β parameter obtained at 0° does not agree with that obtained at 90°; the reason for this is not clear at present. One possibility is that the least-squares fit using only one common β parameter for every kinetic-energy release (KER) component is not an accurate means of simulation. The β parameters measured at the two angles also do not agree with one another at 43 eV probably because of the poor quality of the PIPICO spectra. The background of the PIPICO spectra is wavy due to random coincidences of ions produced in single ionization. This wavy background is added to the PIPICO spectra. In Fig. 4, the mean value of β measured at the two angles is 0.69 at 39 eV, 0.2±0.1 in the region 40–47.5 eV except for the one at 43 eV, about 0.4 in the region 50–60 eV, and gradually decreases above 60 eV.

It has been observed by Lablanquie *et al.* [24] that the dissociative double photoionization ($C^+ + O^+$) takes place with the threshold at 38.4±0.5 eV, which is below the threshold for the molecular double photoionization at 41.3±0.2 eV [25]. They proposed that an indirect process in which highly excited CO^{*+} states autoionize to the double-ionization continuum is responsible for production of the ion pair. Furthermore, Becker *et al.* [26] reported that autoionization of valence-excited CO^{*+} states in the molecule and after dissociation plays a significant role above the double photoionization threshold with emission of a low kinetic-energy electron. At 50 eV, the partial cross section for direct double ionization is comparable to that for autoionization of CO^{*+} [26]. A recent study on the kinetic-energy release distributions of the $C^+ + O^+$ ion pair also indicates that indirect double-photoionization processes are significant in the region 38.4–50 eV [23]. Thus, dissociative double photoionization of CO is complex and the discussion of the observed asymmetry parameter is not straightforward. Theoretical calculations are clearly needed to elucidate the asymmetry parameter of the ion pair.

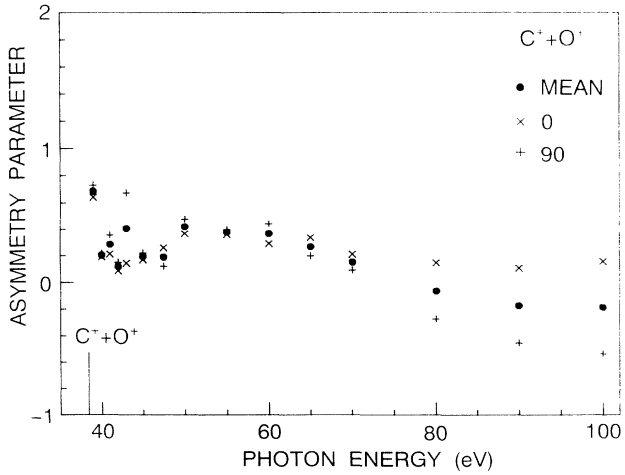

FIG. 4. Asymmetry parameter β for the $C^+ + O^+$ channel of CO^{2+} as a function of excitation energy.

The valence-shell electronic configuration of CO in the ground electronic state is

$$(1\sigma)^2(2\sigma)^2(3\sigma)^2(4\sigma)^2(1\pi)^4(5\sigma)^2:{}^1\Sigma^+$$

in the independent-particle description. In the two-electron transition, the propensity rules may be written as

$$\Delta\lambda_1=0,\pm1\ ,\qquad \Delta\lambda_2=0,\pm1,\pm2 \tag{2}$$

under the restriction of $\Delta\Lambda=0,\pm1$, where λ_1 and λ_2 are the orbital angular momentum of two electrons and Λ is the total angular momentum of electrons. The propensity rules assumed here are different from those in a previous paper [19] and Eq. (2) is considered to be more general than the previous ones. The symmetry of the doubly charged ionic states, those of two photoelectrons, and the total symmetry given by the vector sum of these symmetries of the doubly charged ionic state and two photoelectrons are listed in Table I for the possible electron configuration of CO^{2+}. The value of the β parameter is also listed in Table I on the assumption that the partial cross sections for the individual Π channels are equal to one another and also to that of the Σ channel, as a rough approximation, and by using the equation [2]

$$\beta=\frac{2\sigma_{\parallel}-\sigma_{\perp}}{\sigma_{\parallel}+\sigma_{\perp}}\ , \tag{3}$$

where $\sigma_{\parallel}$ is the partial cross section for the Σ-Σ type transition and $\sigma_{\perp}$ is that for the Σ-Π transition.

If the $C^+ + O^+$ ion pair were produced only from direct double photoionization and subsequent dissociation, Table I indicates that β does not change considerably and is in the range 0.17–0.31. Although direct double ionization is more dominant than the indirect processes above about 60 eV [26], the observed trend of the asymmetry parameter does not follow the simple prediction in Table I. In the case of single photoionization of CO, the oscillator strength for the $3\sigma\rightarrow\varepsilon\sigma$ and $\varepsilon\pi$ ionization have been obtained by theoretical calculations [27], and the results indicate that the oscillator strength for the $3\sigma\rightarrow\varepsilon\sigma$ channel (total symmetry is Σ) is much greater than that for the $3\sigma\rightarrow\varepsilon\pi$ channel (total symmetry is Π) below 55 eV. Above 63 eV, however, the reverse is true [27]. A similar situation may occur in direct double ionization whereby the cross sections for the Σ channels [$\sigma(\Sigma\text{-}\Sigma)$] are greater than those for the Π channels [$\sigma(\Sigma\text{-}\Pi)$] in lower energies and $\sigma(\Sigma\text{-}\Sigma)<\sigma(\Sigma\text{-}\Pi)$ in higher energies. The gradual decrease in the observed β above 60 eV strongly suggests that the intensity alternation in $\sigma(\Sigma\text{-}\Sigma)$ and $\sigma(\Sigma\text{-}\Pi)$ mentioned above occurs in this energy region.

Below the excitation energy of about 41.4 eV, only indirect processes are responsible for the $C^+ + O^+$ ion pair formation via autoionization [24]. CO^{*+} states (Rydberg states converging to the CO^{2+} states) must exist in this region and "configuration-interaction" states (dominant

TABLE I. Symmetry and asymmetry parameter for direct double photoionization of CO.

Electron configuration of CO^{2+}	Symmetry of CO^{2+} ion state	Symmetry of e_1	Symmetry of e_2	Total symmetry	Asymmetry parameter β[a]	
σ^{-2}	Σ	σ	σ,π	Σ,Π	0.20	
		π	σ,π,δ	Π,Σ,Π		
$\sigma^{-1}\pi^{-1}$	Π	σ	σ,π,δ	Π,Σ,Π		
		π	σ,π,δ,ϕ	Σ,Π,Σ,Π		
		σ	σ,π,δ	Π,Σ,Π	0.31	
		π	σ,π,δ	Σ,Π,Σ		
		δ	σ,π,δ	Π,Σ,Π		
π^{-2}	Σ	σ	σ,π	Σ,Π		
		π	σ,π,δ	Π,Σ,Π	0.13	0.17
		δ	π,δ,ϕ	Π,Σ,Π		
	Δ	σ	π,δ,ϕ	Π,Σ,Π		
		π	σ,π,δ,ϕ	Π,Σ,Π,Σ	0.20	
		δ	σ,π,ϕ	Σ,Π,Π		

[a]Under the assumption that the cross sections for the individual Σ and Π channels of the total symmetry are the same.

$3\sigma^{-1}\,^2\Sigma^+$ states) have also been observed there by photoelectron spectroscopy [28–31]. Furthermore, CO** states (double Rydberg states converging to the CO^{2+} states) may contribute to the ion pair formation [23]. The observed β is 0.69 at the lowest excitation energy, 39 eV, and then decreases to 0.20–0.28 in the region 40–41 eV, which means that the total symmetries involved in the transition changes considerably in this region. If autoionization takes place before dissociation, the final CO^{2+} state is likely the repulsive $1\,^3\Sigma^-(1\pi^{-2})$ state.

The method to predict the asymmetry parameter in direct double ionization is now used for the case of indirect double ionization of CO*+ and CO**. The angular distributions of ion pairs will follow the total symmetry of the states populated immediately after single photon absorption, as is the case for inner-shell excitation and ionization. The results are shown in Tables II and III for CO*+ and CO**, respectively. In the case of indirect double photoionization via CO*+, β does not change very much, if β is taken together for the CO*+

TABLE II. Symmetry and asymmetry parameter for indirect double photoionization of CO via CO*+.

Electron configuration of CO*+	Electron configuration to which CO*+ states converge	Symmetry of CO*+ state	Symmetry of photoelectron	Total symmetry	Asymmetry parameter β[a]	
$\sigma^{-1}(m\sigma\rightarrow n\sigma)$	σ^{-2}	Σ	σ,π	Σ,Π	0.5	
$\sigma^{-1}(m\sigma\rightarrow n\pi)$		Π	σ,π,δ	Π,Σ,Π	0	0
$\sigma^{-1}(m\sigma\rightarrow n\delta)$		Δ	π	Π	-1	
$\sigma^{-1}(1\pi\rightarrow n\sigma)$	$\sigma^{-1}\pi^{-1}$	Π	σ,π,δ	Π,Σ,Π	0	
$\sigma^{-1}(1\pi\rightarrow n\pi)$		Σ	σ,π	Σ,Π	0.5	
		Δ	π,δ	Π,Σ		
$\sigma^{-1}(1\pi\rightarrow n\delta)$		Π	σ,π,δ	Π,Σ,Π	-0.25	
		Φ	δ	Π		
$\sigma^{-1}(1\pi\rightarrow n\phi)$		Σ	σ,π	Σ,Π	0	0.04
		Δ	π	Π		
$\pi^{-1}(m\sigma\rightarrow n\sigma)$		Π	σ,π,δ	Π,Σ,Π	0	
$\pi^{-1}(m\sigma\rightarrow n\pi)$		Σ	σ,π	Σ,Π	0.2	
		Δ	π,δ,ϕ	Π,Σ,Π		
$\pi^{-1}(m\sigma\rightarrow n\delta)$		Π	σ,π,δ	Π,Σ,Π	-0.25	
		Φ	δ	Π		
$\pi^{-1}(1\pi\rightarrow n\sigma)$	π^{-2}	Σ	σ,π	Σ,Π	0.25	
		Δ	π,δ,ϕ	Π,Σ,Π		
$\pi^{-1}(1\pi\rightarrow n\pi)$		Π	σ,π,δ	Π,Σ,Π	0.25	
		Φ	δ,ϕ	Π,Σ		0.11
$\pi^{-1}(1\pi\rightarrow n\delta)$		Σ	σ,π	Σ,Π		
		Δ	π,δ,ϕ	Π,Σ,Π	0	
		Γ	ϕ	Π		
$\pi^{-1}(1\pi\rightarrow n\phi)$		Π	σ,π,δ	Π,Σ,Π	0	

[a]Under the assumption that the cross sections for the individual Σ and Π channels of the total symmetry are the same.

states converging to the same CO^{2+} electron configuration (shown by large braces in Table II). In the case of indirect double photoionization via CO^{**}, β is rather large for the CO^{**} states converging to the $\sigma^{-1}\pi^{-1}$ and π^{-2} CO^{2+} configurations (Table III). These CO^{**} states may be strong candidates for yielding a β value of 0.69 at 39 eV. However, since calculations [27] show that the $3\sigma \rightarrow \varepsilon\sigma$ channel of CO is about three or four times more intense than the $3\sigma \rightarrow \varepsilon\pi$ channel at certain energies and a similar situation may occur in indirect double ionization, the channels having a β value larger than -0.25 may also be candidates for indirect double ionization at 39 eV. It is unfortunate that definite candidates cannot be determined for indirect double ionization from the observed β at 39 eV. As listed in Tables I–III, there are so many channels contributing to the $C^+ + O^+$ ion pair production that further discussion of the β parameter at other excitation energies may be meaningless without the help of theoretical calculations.

The ratio of the cross section for the parallel transition

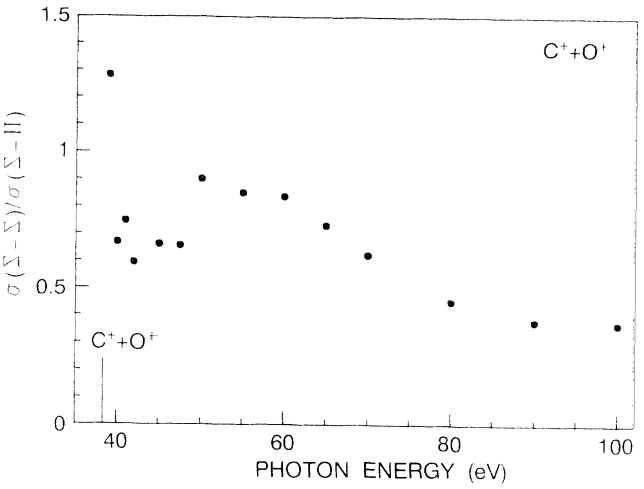

FIG. 5. The ratios of cross sections $\sigma(\Sigma\text{-}\Sigma)/\sigma(\Sigma\text{-}\Pi)$ for the $C^+ + O^+$ channel of CO^{2+} as a function of excitation energy.

TABLE III. Symmetry and asymmetry parameter for indirect double photoionization of CO via CO^{**}.

Electron configuration of CO^{**}	Electron configuration to which CO^{**} states converge	Symmetry of CO^{**} state	Asymmetry parameter β[a]	
$(k\sigma \rightarrow l\sigma)(m\sigma \rightarrow n\sigma)$	σ^{-2}	Σ	0.5	
$(m\sigma \rightarrow n\pi)$		Π		
$(k\sigma \rightarrow l\pi)(m\sigma \rightarrow n\sigma)$		Π		0
$(m\sigma \rightarrow n\pi)$		Σ	0	
$(m\sigma \rightarrow n\delta)$		Π		
$(k\sigma \rightarrow l\delta)(m\sigma \rightarrow n\pi)$		Π	-1	
$(k\sigma \rightarrow l\sigma)(1\pi \rightarrow n\sigma)$	$\sigma^{-1}\pi^{-1}$	Π		
$(1\pi \rightarrow n\pi)$		Σ	0.5	
$(1\pi \rightarrow n\delta)$		Π		
$(1\pi \rightarrow n\phi)$		Σ		
$(k\sigma \rightarrow lpi)(1\pi \rightarrow n\sigma)$		Σ		
$(1\pi \rightarrow n\pi)$		Π	0.5	0.36
$(1\pi \rightarrow n\delta)$		Σ		
$(1\pi \rightarrow n\phi)$		Π		
$(k\sigma \rightarrow l\delta)(1\pi \rightarrow n\sigma)$		Π	0	
$(1\pi \rightarrow n\pi)$		Σ		
$(1\pi \rightarrow n\delta)$		Π		
$(1\pi \rightarrow l\sigma)(1\pi \rightarrow n\sigma)$	π^{-2}	Σ	0.5	
$(1\pi \rightarrow n\pi)$		Π		
$(1\pi \rightarrow n\delta)$		Σ		
$(1\pi \rightarrow n\phi)$		Π		
$(1\pi \rightarrow l\pi)(1\pi \rightarrow n\sigma)$		Π		
$(1\pi \rightarrow n\pi)$		Σ	0.5	
$(1\pi \rightarrow n\delta)$		Π		0.4
$(1\pi \rightarrow n\phi)$		Σ		
$(1\pi \rightarrow l\delta)(1\pi \rightarrow n\sigma)$		Σ		
$(1\pi \rightarrow n\pi)$		Σ	0.5	
$(1\pi \rightarrow n\delta)$		Π		
$(1\pi \rightarrow n\phi)$		Σ		
$(1\pi \rightarrow l\phi)(1\pi \rightarrow n\sigma)$		Π	0	
$(1\pi \rightarrow n\pi)$		Σ		
$(1\pi \rightarrow n\delta)$		Π		

[a]Under the assumption that the cross sections for the individual Σ and Π channels of the total symmetry are the same.

to that for the vertical transition, which is given by $(1+\beta)/(2-\beta)$, is shown in Fig. 5 as a function of excitation energy. The mean values of the β parameter in Fig. 4 were taken and the one at 43 eV was omitted.

IV. CONCLUSIONS

Angle-resolved photoion-photoion coincidence spectra have been measured for double photoionization of CO in the energy region of valence orbitals by use of linearly polarized synchrotron radiation. The results indicate clearly that anisotropies exist for the $C^+ + O^+$ dissociation channel of CO^{2+}. Although the observed anisotropies must be related to the symmetry-characterized dissociation, because of the presence of so many channels contributing to the ion pair formation even at low excitation energies, the trend in the observed β parameter cannot be explained by symmetry considerations involved in the transitions. In this regard, theoretical calculations are clearly needed to elucidate the asymmetry parameter of the ion pair. From the observed β parameter, the ratio of the cross section for the parallel transition to that for the vertical transition has been determined in the excitation energy region of 39–100 eV.

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