Nondipole effects in the photoionization of neon: Random-phase approximation

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The random-phase approximation (RPA) is applied to study nondipole corrections to the angular distribution of photoelectrons from the $n=2$ shell of Ne. Calculations of the parameters γ_{nl} and δ_{nl} arising from E_1-E_2 interference effects are carried out for the $2s$ and $2p$ subshells of Ne in the photon energy range 100–2000 eV. For the $2s$ shell, the RPA calculations show small effects of correlation near the $2s$ threshold energy, but are otherwise in agreement with independent-particle approximation (IPA) calculations. The RPA and IPA values of γ_{2s} are also in agreement with experiment. For the $2p$ shell, a small difference between RPA and IPA calculations of the nondipole parameters is found for energies near the $1s$ threshold; however, both RPA and IPA calculations of the parameter $\gamma_{2p}+3\delta_{2p}$ disagree significantly with experimental measurements for photon energies above 1000 eV. [S1050-2947(99)09605-5]

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I. INTRODUCTION

Recent measurements of photoionization of closed-shell atoms for photon energies in the range 100–5000 eV [1–4] convincingly demonstrate the breakdown of the dipole approximation and provide sufficient quantitative data to initiate detailed new theoretical investigations of photoelectron angular distributions in regions where the dipole approximation is no longer valid.

Theoretical studies of nondipole effects in photoionization of multielectron atoms, with allowance for electron correlation, appeared more than twenty years ago in the work of Amusia *et al.* [5]. Since then, many investigations of effects beyond the dipole approximation have been carried out, including studies of low-energy dipole and quadrupole autoionizing resonances in the outer-shell photoionization of Ar and Mn [6]. Relativistic studies of dipole-quadrupole interference corrections to the photoelectron angular distribution for $1s$, $2s$, and $2p$ subshells of atoms with nuclear charges Z ranging from 6 to 40, carried out using Coulomb-field and screened Coulomb-field approximations, were presented by Pratt and Bechler in Ref. [7]. General formulas for the interference contributions to the differential cross section from higher multipoles in relativistic calculations were given by Scofield in Ref. [8], where detailed numerical calculations for Ne-like Ba and He-like Ni were carried out in the relativistic independent-particle approximation (IPA) using a Dirac-Slater central potential. Extensive nonrelativistic numerical results for the nondipole asymmetry parameters for inner subshells of all noble-gas atoms from He to Xe, obtained in the IPA using a Hartree-Slater potential, were presented more recently by Cooper [9].

Very recently, a breakdown of the IPA in the dipole $2p$ photoionization of Ne far above threshold (200–1400 eV) was demonstrated [10], prompting a search for the same phenomenon in the E_1-E_2 interference spectrum of Ne. In Ref. [3], it was found that the experimental data for the Ne nondipole angular distribution asymmetry parameter were as much as 50% higher than IPA predictions far above threshold (200–1400 eV), indicating a significant effect of electron correlation on the nondipole parameters in the keV photon energy region.

These developments have prompted us to extend both the relativistic random-phase approximation (RRPA) [11] and the nonrelativistic random-phase approximation with exchange (RPAE) [12] beyond the dipole approximation to investigate in detail the dipole-quadrupole interference effects in the keV photon energy region in general, and to apply the newly developed methods to the Ne $2p$ photoionization, to interpret the recent experimental data from Ref. [3].

In the following section, we present numerical results from RPA calculations and make comparisons with the previous IPA calculations and with existing experimental data. We then give a general account of a possible role of electron correlation in Ne for the nondipole angular distribution parameter $\zeta_{2p}=\gamma_{2p}+3\delta_{2p}$, which was measured in Ref. [3], and for the dipole parameter β_{2p}. We show that the latter is significantly altered by electron correlation far above threshold, whereas the former is insensitive to multielectron effects in this case. The relativistic and nonrelativistic formulas used to study E_1-E_2 contributions to the nondipole photoelectron angular distribution parameters are collected in the Appendixes.

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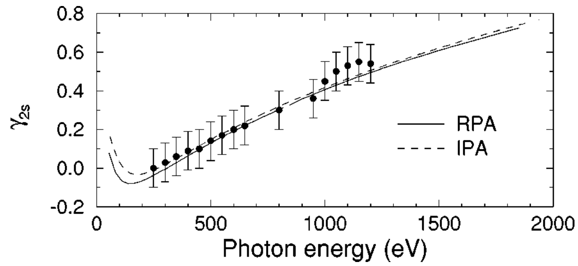

FIG. 1. Asymmetry parameter γ_{2s} for the $2s$ subshell of Ne is compared with IPA calculations from [9] and with experimental data from [3].

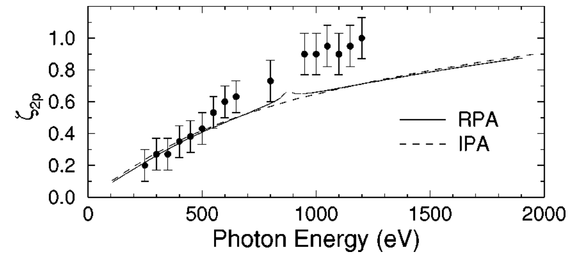

FIG. 2. Asymmetry parameter $\gamma_{2p}+3\delta_{2p}$ for the $2p$ subshell of Ne is compared with IPA calculations from [9] and with experimental data from [3].

II. RESULTS AND DISCUSSION

The differential cross section for photoionization of an electron from subshell (n,κ) of a closed-shell atom may be written in the form given in Ref. [9]:

$$\frac{d\sigma_{n\kappa}}{d\Omega}=\frac{\sigma_{n\kappa}}{4\pi}[1+\beta_{n\kappa}P_2(\cos\theta)+(\delta_{n\kappa}+\gamma_{n\kappa}\cos^2\theta)\sin\theta\cos\phi], \tag{1}$$

where θ and ϕ are the polar angles of the electron momentum vector in a coordinate system with the polarization vector $\hat{\epsilon}$ directed along the z axis and the photon propagation vector $\hat{k}$ directed along the x axis. In the dipole approximation, the parameter $\beta_{n\kappa}$ characterizes the photoelectron angular distribution completely. The two parameters $\delta_{n\kappa}$ and $\gamma_{n\kappa}$ describe the leading corrections beyond the dipole approximation, which arise from a combination of E_1-E_2 and E_1-M_1 interference effects. The E_1-E_2 contributions are proportional to the photon momentum k, for small values of k. The relativistic M_1 photoionization amplitudes vanish in the Pauli approximation and are found to be insignificant numerically; the nonrelativistic M_1 amplitudes vanish identically. We therefore include only E_1-E_2 contributions to $\delta_{n\kappa}$ and $\gamma_{n\kappa}$ in the present studies.

In our RPA calculations of the E_1 photoionization amplitudes for Ne, all excitations from $1s$ and $2s$ shells to continuum p states, and from the $2p$ shell to continuum s and d states, are included. This leads to a coupled four-channel problem nonrelativistically and a coupled nine-channel problem relativistically. Similarly, the RPA calculation of E_2 amplitudes leads to a four-channel nonrelativistic or a ten-channel relativistic problem, in which $1s$ and $2s$ shells are excited to continuum d states and the $2p$ shell is excited to continuum p and f states. It should be noted that dipole and quadrupole transition amplitudes calculated using either RPAE or RRPA are independent of gauge, so length-form and velocity-form amplitudes are identical. The numerical results from the present RRPA and RPAE calculations are virtually indistinguishable. We present the RRPA results in our figures, but use the simpler RPAE theory for qualitative discussions of our results.

Since $\delta_{n\kappa}=0$ for s subshells, nonrelativistically, in Fig. 1 we compare fully coupled RPA calculations of the remaining nondipole parameter γ_{2s} with IPA calculations [9] and with experimental results from [3]. The RPA and IPA calculations are seen to be in close agreement with each other except near the $2s$ threshold, where a noticeable effect of electron correlation on the RPA parameter is seen. Both calculations agree with the experimental measurements [3] for this case. In Fig. 2 we compare the present RPA values of the parameter $\zeta_{2p}=\gamma_{2p}+3\delta_{2p}$ for Ne with the IPA calculations of [9] and the experimental data of [3]. The only appreciable difference between the RPA and IPA calculations is the small feature in the RPA ζ_{2p} parameter near the $1s$ threshold, caused by intershell coupling. In this case, however, there is a substantial difference between the RPA calculations and experiment. This is all the more puzzling against the background of the Ne dipole β_{2p}-parameter spectrum, for which the experimental and RPA results [3] are in excellent agreement with each other and where both differ substantially from the IPA calculations.

In the following paragraphs, we examine the sensitivity of both ζ_{2p} and β_{2p} to electron correlation effects and find that, indeed, one should not expect substantial deviations of ζ_{2p} from the IPA results in the high-energy region, in contrast to β_{2p}.

A. The Ne ζ_{2p}-parameter spectrum

For $2p$ subshells, the expression for ζ_{2p} is given in Appendix B. We first comment that the dipole amplitude for $l\to l+1$ is much larger than that for $l\to l-1$ $(D_2\gg D_0)$; correspondingly, the quadrupole amplitude for $l\to l+2$ is much larger than that for $l\to l$ $(Q_3\gg Q_1)$ over the energy range considered. It follows that the expression for ζ_{2p} given in Eq. (B14) can be well approximated by its leading term,

$$\zeta_{2p}\approx\frac{7k}{\sqrt{10}}\rho_{qd}\cos(\delta_3-\delta_2), \tag{2}$$

where $\rho_{qd}=Q_3/D_2$.

We note first that $\cos(\delta_3-\delta_2)$ is close to unity. It depends on differences between phase shifts with large values of l $(l=2$ and $l=3)$, which, for low Z atoms and hence for Ne, must be very small at high energies. Indeed,

$$\delta_3-\delta_2=(\tilde{\delta}_3-\tilde{\delta}_2)+(\delta_3^C-\delta_2^C). \tag{3}$$

The latter term, the difference between Coulomb phase shifts δ_l^C, vanishes at high energies [13], and can thus be neglected in the present consideration. The non-Coulomb phase shifts $\tilde{\delta}_l$ with $l=2$ and 3 for low Z atoms are known to vanish at threshold and, although they increase somewhat with energy before eventually decreasing at high energies, the difference between them remains quite small in the photon energy region under discussion. Thus, from general considerations, $\delta_3-\delta_2$ is close to zero and $\cos(\delta_3-\delta_2)$ is close to unity. Our calculated IPA and RPA results, in full support of this conclusion, show that for Ne $2p$ photoionization $\cos(\delta_3-\delta_2)$ varied between 0.95 and 0.98 over a broad photon energy region from 1000 to 4000 eV.

Electron correlation can influence the ζ_{2p} parameter through either the ratio ρ_{qd} or through $\cos(\delta_3-\delta_2)$. However, as was shown above, $\cos(\delta_3-\delta_2)$ is near its maximum, and is, therefore, insensitive to small changes of its argument; moreover, large changes in its argument are not possible for photon energies above all ionization thresholds. We can therefore conclude that the ζ_{2p} parameter cannot be significantly altered by electron correlation through variation of $\cos(\delta_3-\delta_2)$. The ζ_{2p} parameter is not affected significantly through ρ_{qd} either. This quantity depends on the dominant $l\rightarrow l+1$ dipole and $l\rightarrow l+2$ quadrupole photoionization amplitudes, which are not normally sensitive to electron correlation. Furthermore, ρ_{qd} is a ratio, and thus possible correlation contributions to each of the matrix elements can mutually cancel. In fact, our calculations show only a tiny change in ρ_{qd}, brought about by intershell correlation for photon energies near the $1s$ threshold.

We thus find that ζ_{2p} is insensitive to electron correlation, in agreement with our numerical calculations. Based on this conclusion, the approximately 50% difference between experimental data [3] and calculated RPA or IPA values for the Ne ζ_{2p} parameter is difficult to understand; either correlation effects beyond the RPA theory are important, which is hard to believe at the high energies under discussion, or there is some unknown systematic error in the experiment.

B. The Ne β_{2p}-parameter spectrum

In contrast to the ζ_{2p} parameter considered above, appreciable (about 30%) differences between IPA and RPA calculations are found for the Ne β_{2p} parameter. These differences are brought about by the electron correlation. To understand this, we note that under the assumption $D_2 \gg D_0$, we may approximate Eq. (B11) of Appendix B as

$$\beta_{2p} \approx 1 + 2\sqrt{2}\rho_0 \cos(\delta_2-\delta_0), \tag{4}$$

where $\rho_0 = D_0/D_2$. As to the sensitivity of $\cos(\delta_2-\delta_0)$ to electron correlation, using the same analysis as above, it is easy to see that whereas δ_2 is negligible, δ_0 still retains appreciable values in the range of photon energies under discussion. We find that, in this energy region, $\delta_2-\delta_0$ is close to $\pi/2$, so that $\cos(\delta_2-\delta_0)$ is near zero. Hence, small changes in the phase shift difference can lead to large changes in $\cos(\delta_2-\delta_0)$ and, thereby, in the β_{np} parameter. The β_{np} parameter can be affected through ρ_0 as well, because the weaker amplitude D_0 is more sensitive to electron correlation than the stronger amplitude D_2. Therefore, the parameter ρ_0 is also more sensitive to electron correlation than the ratio ρ_{qd} discussed in the preceding subsection. From these arguments, we conclude that β_{np} should be much more sensitive to the electron correlation than the nondipole parameter ζ_{np}.

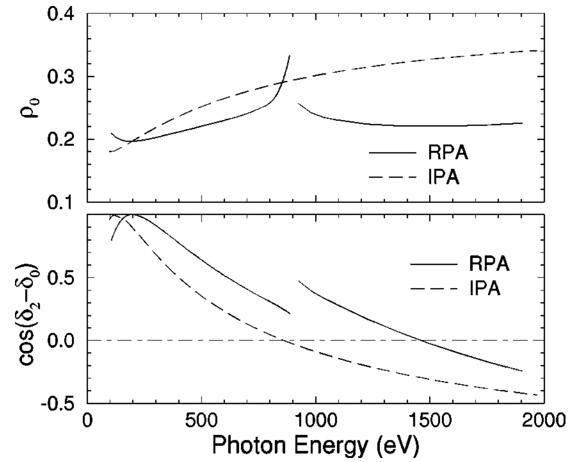

FIG. 3. RPA and IPA calculations of the quantities $\cos(\delta_0-\delta_2)$ and ρ_0 are compared.

Our calculated IPA and RPA results for $\cos(\delta_2-\delta_0)$ and for ρ_0 are displayed in Fig. 3. One can see that, in a broad energy region, the IPA values of $\cos(\delta_2-\delta_0)$ are indeed small. Hence, as follows from the discussion above, $\cos(\delta_2-\delta_0)$ is very sensitive to electron correlation effects, leading to significant differences between IPA and RPA values of $\cos(\delta_2-\delta_0)$ shown in Fig. 3. The IPA and RPA values even have opposite signs in a broad energy region above the $1s$-ionization threshold. Thus, the second term in Eq. (4) changes sign under the action of correlation, leading to substantial differences between IPA and RPA values of the Ne β_{2p}-parameter spectrum.

III. CONCLUSION

In conclusion, we have presented a detailed discussion of properties of the Ne β_{2p}- and ζ_{2p}-parameter spectra. We have seen that ζ_{2p} is insensitive to the influence of electron correlation far above threshold, in contrast to β_{2p}. Based on this, the $\approx$50% difference between the IPA and experimental data for the ζ_{2p}-parameter spectrum of Ne at high energies, reported in Ref. [3], is difficult to understand, at least within the framework of RPA. Further theoretical and experimental investigations of this spectrum are clearly required.

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APPENDIX A: RELATIVISTIC FORMULAS

The cross section $\sigma_{n_b\kappa_b}$ for photoionization of an electron from subshell (n_b,κ_b) of a closed-shell atom is given by

$$\sigma_{n_b\kappa_b}=\frac{4\pi^2\alpha}{3}\omega\sum_{\kappa}|D_{\kappa\kappa_b}|^2, \tag{A1}$$

where $D_{\kappa\kappa_b}$ is the reduced dipole matrix element

$$D_{\kappa\kappa_b}=i^{1-l}e^{i\delta_\kappa}\langle\epsilon\kappa||q_1^{(1)}||n_b\kappa_b\rangle. \tag{A2}$$

The operator $q_1^{(1)}$ is the electric dipole transition operator defined in [11] and δ_κ is the phase shift of the outgoing electron.[1] The dipole angular distribution asymmetry parameter $\beta_{n_b\kappa_b}$ is given by

$$\beta_{n_b\kappa_b}=\left[\sum_{\kappa\kappa'}\mathcal{B}(j,j';j_b)D_{\kappa\kappa_b}D^*_{\kappa'\kappa_b}\right]\left[\sum_{\kappa}|D_{\kappa\kappa_b}|^2\right]^{-1}, \tag{A3}$$

with

$$\mathcal{B}(j,j';j_b)=\sqrt{30}(-1)^{j'+j_b}\langle j'||C_2||j\rangle\begin{Bmatrix}1 & 1 & 2\\ j & j' & j_b\end{Bmatrix}. \tag{A4}$$

The coefficients $\mathcal{B}(j,j';j_b)$ are symmetric with respect to interchange of the first pair of arguments.

We write the nondipole asymmetry parameters $\delta_{n\kappa}$ and $\gamma_{n\kappa}$ in terms of two auxiliary parameters $\Gamma^1_{n_b\kappa_b}$ and $\Gamma^3_{n_b\kappa_b}$:

$$\Gamma^i_{n_b\kappa_b}=k\left[\sum_{\kappa'\kappa}\mathcal{D}_i(j,j';j_b)\,\mathrm{Im}(D_{\kappa\kappa_b}Q^*_{\kappa'\kappa_b})\right]\left[\sum_{\kappa}|D_{\kappa\kappa_b}|^2\right]^{-1}, \tag{A5}$$

for $i=1$ or 3. In these expressions,

$$Q_{\kappa\kappa_b}=i^{1-l}e^{i\delta_\kappa}\langle\epsilon\kappa||q_2^{(1)}||n_b\kappa_b\rangle$$

is the reduced matrix element of the electric quadrupole operator $q_2^{(1)}$, and

$$\mathcal{D}_1(j,j';j_b)=\sqrt{\frac{27}{10}}(-1)^{j'+j_b}\langle j'||C_1||j\rangle\begin{Bmatrix}2 & 1 & 1\\ j & j' & j_b\end{Bmatrix}, \tag{A6}$$

$$\mathcal{D}_3(j,j';j_b)\sqrt{\frac{21}{5}}(-1)^{j'+j_b}\langle j'||C_3||j\rangle\begin{Bmatrix}2 & 1 & 3\\ j & j' & j_b\end{Bmatrix}. \tag{A7}$$

The differential cross section for unpolarized incident radiation may be expressed in terms of the parameters $\beta_{n_b\kappa_b}$, $\Gamma^1_{n_b\kappa_b}$, and $\Gamma^3_{n_b\kappa_b}$ as

$$\frac{d\sigma_{n_b\kappa_b}}{d\Omega}=\frac{\sigma_{n_b\kappa_b}}{4\pi}\left[1-\frac{1}{2}\beta_{n_b\kappa_b}P_2(\cos\psi)+\Gamma^1_{n_b\kappa_b}P_1(\cos\psi)+\Gamma^3_{n_b\kappa_b}P_3(\cos\psi)\right], \tag{A8}$$

where ψ is the angle between the photon propagation direction $\hat{k}$ and the direction of the photoelectron $\hat{p}$. For linearly polarized radiation, the differential cross section is given by Eq. (1) with

$$\delta_{n_b\kappa_b}=\Gamma^1_{n_b\kappa_b}+\Gamma^3_{n_b\kappa_b}, \tag{A9}$$

$$\gamma_{n_b\kappa_b}=-5\Gamma^3_{n_b\kappa_b}. \tag{A10}$$

APPENDIX B: NONRELATIVISTIC FORMULAS

A nonrelativistic approach significantly facilitates theoretical discussion, and, for light atoms such as Ne, where relativistic (fine-structure) corrections to wave functions are small, leads to results in close agreement with the relativistic theory. Indeed, the nonrelativistic formulas can be written in precisely the same form as the relativistic formulas with several modifications: The nonrelativistic expression for the cross section $\sigma_{n_bl_b}$ is

$$\sigma_{n_bl_b}=\frac{8\pi^2\alpha}{3}\omega\sum_{\kappa}|D_{ll_b}|^2, \tag{B1}$$

where D_{ll_b} is the reduced dipole matrix element

$$D_{ll_b}=i^{-l}e^{i\delta_l}\langle\epsilon l||q_1^{(1)}||n_bl_b\rangle. \tag{B2}$$

The dipole angular distribution asymmetry parameter $\beta_{n_bl_b}$ is given by

$$\beta_{n_bl_b}=\left[\sum_{ll'}\mathcal{B}(l,l';l_b)D_{ll_b}D^*_{l'l_b}\right]\left[\sum_{l}|D_{ll_b}|^2\right]^{-1}, \tag{B3}$$

with

$$\mathcal{B}(l,l';l_b)=\sqrt{30}(-1)^{l'+l_b}\langle l'||C_2||l\rangle\begin{Bmatrix}1 & 1 & 2\\ l & l' & l_b\end{Bmatrix}. \tag{B4}$$

The coefficients $\mathcal{B}(l,l';l_b)$ are symmetric with respect to interchange of the first pair of arguments.

Again, we introduce two auxiliary parameters $\Gamma^1_{n_bl_b}$ and $\Gamma^3_{n_bl_b}$:

$$\Gamma^i_{n_b\kappa_b}=k\left[\sum_{\kappa'\kappa}\mathcal{D}_i(j,j';j_b)\,\mathrm{Im}(D_{ll_b}Q^*_{l'l_b})\right]\left[\sum_{l}|D_{ll_b}|^2\right]^{-1}, \tag{B5}$$

for $i=1$ or 3. In these expressions,

$$Q_{ll_b}=i^{-l}e^{i\delta_l}\langle\epsilon l||q_2^{(1)}||n_bl_b\rangle$$

is the reduced matrix element of the electric quadrupole operator $q_2^{(1)}$, and

$$\mathcal{D}_1(l,l';l_b)=\sqrt{\frac{27}{10}}(-1)^{l'+l_b}\langle l'||C_1||l\rangle\begin{Bmatrix}2 & 1 & 1\\ l & l' & l_b\end{Bmatrix}, \tag{B6}$$

[1]We use the conventional definition of reduced matrix element here, leading to a phase difference of $(-1)^{j-j_b}$ with [11].

$$\mathcal{D}_3(l,l';l_b)=\sqrt{\frac{21}{5}}(-1)^{l'+l_b}\langle l'||C_3||l\rangle\begin{Bmatrix}2 & 1 & 3\\ l & l' & l_b\end{Bmatrix}. \tag{B7}$$

Again, as in the relativistic case, $\delta_{n_b l_b}=\Gamma^1_{n_b l_b}+\Gamma^3_{n_b l_b}$ and $\gamma_{n_b l_b}=-5\Gamma^3_{n_b l_b}$. The present formulas for the nondipole parameters agree with those given in Refs. [5,9]; however, there are several misprints in Table XII of Ref. [9].

For the $2s$ subshell, the angular distribution parameters are

$$\beta_{2s}=2, \tag{B8}$$

$$\delta_{2s}=0, \tag{B9}$$

$$\gamma_{2s}=3k\frac{Q_2}{D_1}\cos(\delta_2-\delta_1), \tag{B10}$$

where $D_1\equiv|D_{1\,2s}|$ and $Q_2\equiv|Q_{2\,2s}|$. For the $2p$ subshell, we have

$$\beta_{2p}=\frac{1}{\overline{\sigma}_{2p}}[2\sqrt{2}D_0D_2\cos\Delta_{20}+D_2^2], \tag{B11}$$

$$\delta_{2p}=\frac{k}{10\overline{\sigma}_{2p}}[\sqrt{30}D_0Q_1\cos\Delta_{10}-\sqrt{15}D_2Q_1\cos\Delta_{21}+\sqrt{10}D_2Q_3\cos\Delta_{32}-2\sqrt{5}D_0Q_3\cos\Delta_{30}], \tag{B12}$$

$$\gamma_{2p}=\frac{k}{10\overline{\sigma}_{2p}}[6\sqrt{15}D_2Q_1\cos\Delta_{21}+4\sqrt{10}D_2Q_3\cos\Delta_{32}+10\sqrt{5}D_0Q_3\cos\Delta_{30}], \tag{B13}$$

$$\zeta_{2p}=\frac{k}{10\overline{\sigma}_{2p}}[3\sqrt{30}D_0Q_1\cos\Delta_{10}+3\sqrt{15}D_2Q_1\cos\Delta_{21}+7\sqrt{10}D_2Q_3\cos\Delta_{32}+4\sqrt{5}D_0Q_3\cos\Delta_{30}], \tag{B14}$$

where $\Delta_{ij}=\cos(\delta_i-\delta_j)$ and where $\overline{\sigma}_{2p}=D_0^2+D_2^2$. In these equations, $D_0\equiv|D_{0\,2p}|$, $D_2\equiv|D_{2\,2p}|$, $Q_1\equiv|Q_{1\,2p}|$, and $Q_3\equiv|Q_{3\,2p}|$. The quantity $\zeta_{2p}=\gamma_{2p}+3\delta_{2p}$ is the nondipole parameter measured in Ref. [3].

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