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# Multipole effects in the excitation of high-$l$ doubly excited states in Sr

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We report the observation of unexpected line strengths in the resonant multiphoton laser excitation of high-angular-momentum $7d_{5/2}n'l'$ and $8p_{3/2}n''l''(n''\neq 16)$ states in Sr. The results differ from expected isolated-core excitation features usually explained on the basis of the multichannel-quantum-defect theory. In the present case, the observed line strengths and, in particular, the disappearance of single resonances can be explained by a configuration-interaction calculation, giving explicitly the multipole character of the correlation between the electrons. The spectra offer a sensitive way to test calculations involving electronic correlation.

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Resonant multiphoton laser excitation of quasi-two-electron systems such as the alkaline-earth atoms has been used to investigate the dynamics of two highly excited electrons in the field of a doubly charged core [1–12]. The large spatial volume available to the two electrons allows for pronounced electron correlation effects, which can be traced in absorption spectra almost up to the three-body particle breakup threshold. The relevance of these investigations stems from the close relation to the fundamental three-body Coulomb problem, where only a limited set of experimental data on pure three-body Coulomb systems such as He and $H^-$ exists so far.

Efficient preparation of doubly excited electrons relies on the isolated-core excitation (ICE) technique [1,2,12]. Briefly, in a first step one of the two valence electrons is excited to a Rydberg state, where it is shelved from the interaction with further laser photons. In a second step, a further laser photon, resonant or nearly resonant with a dipole-allowed core transition, acts only on the inner electron. The outer Rydberg electron is shaken into another Rydberg orbit as a result of the change of the core state (monopole interaction) giving rise to shake satellites. So far correlation effects play only a minor role.

Extension of the ICE scheme by resonant multiphoton excitation of the core electron allows for the preparation of asymmetrically excited states with high quantum numbers $NLnl$, where $N<n$. As a result of the large orbits and the high angular momenta for both electrons strong long-range correlation effects lead to substantial deviations from what is expected in a simple single-step ICE scheme and to distinct features observable in absorption spectra.

In particular, the polarization of the inner electron through the outer electron becomes more and more important for higher excited states of the inner electron [4–7,9]. Additional structures in the spectra appear in the vicinity of dipole-forbidden ionic core transitions, indicating the influence of the electric field of the outer electron on the inner one. The effect has been described approximately by a generalized ICE scheme within the multichannel-quantum-defect theory (MQDT) [4,5]. More recently, the influence of the polarized core state on the outer electron has been observed, underlining the dipole structure of doubly excited states [3,13–15].

In this paper, we report the observation of doubly excited Rydberg series in Sr that show characteristic oscillator strength distribution manifested through sharply localized intensity modulations. The familiar shake satellite structure expected from a single-step ICE scheme is largely absent. The modulations do not originate from nearly degenerate perturber states. This situation is somewhat unusual in a MQDT picture, where modulations in the spectra are usually induced through local perturber states and may lead to vanishing linewidths as a result of interferences in the channel interaction [16]. We show that a configuration-interaction (CI) calculation leads to direct identification of the states responsible for the perturbation and allows us to determine precisely the multipole character of the electron-electron interaction. It shows that the present situation is complicated due to the fact that channel interactions play a role even below the energy of the lowest bound state, a rather atypical situation that may not be easily treated by MQDT. Most remarkable is the vanishing of particular lines as a result of a quasiselection rule for the dipole correlation matrix elements.

The apparatus and the resonant multiphoton laser excitation method have been described elsewhere [3]. In brief, strontium atoms in the ground state are excited to a Stark level $5s16n_1$ $(m=0)$ in the presence of a constant electric field by two excimer-laser pumped dye lasers. After the excitation, the electric field is switched off adiabatically within 2.8 $\mu$s and the Stark state is converted to $5s16l$ [17]. Electric stray fields are reduced and estimated to be lower than 50 mV/cm, thus avoiding effectively the mixing between different $l$ states in the manifold. The atoms in the $5s16l$ state are further excited to the $5d_{5/2}16l(l=14,15)$ via the $5p_{3/2}16l$ state by another two dye lasers (linearly polarized). A fifth laser is scanned in the vicinity of the energy range of the $7d_{5/2}n'l'$ and $8p_{3/2}n''l''$ series. This situation is depicted in a level diagram in Fig. 1. The doubly excited atoms either autoionize or, alternately, are directly photoionized by the strong fifth dye laser. The resulting excited $Sr^+$ ions are further photo or pulsed-field ionized to yield $Sr^{2+}$ ions, which

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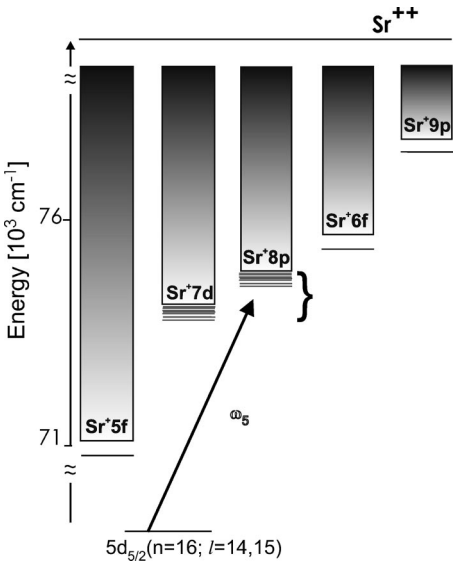


FIG. 1. Shown is a level diagram of the relevant channels. The fifth laser is scanned in the vicinity of the $7dnl$ and $8pnl$ Rydberg series. We note that the fine-structure splitting of the core levels is too small to be resolved. The $n=16$ states of nearby Rydberg series with other core configurations, which act as perturber states, are indicated by horizontal bars. The energy is given with respect to the first ionization threshold.

are detected in our experiments. The absorption spectra are obtained by recording the $Sr^{2+}$ ion yield as a function of the wavelength of the fifth laser. The pulsed field serves also in our time-of-flight spectrometer to sweep the $Sr^{2+}$ ions to the detector.

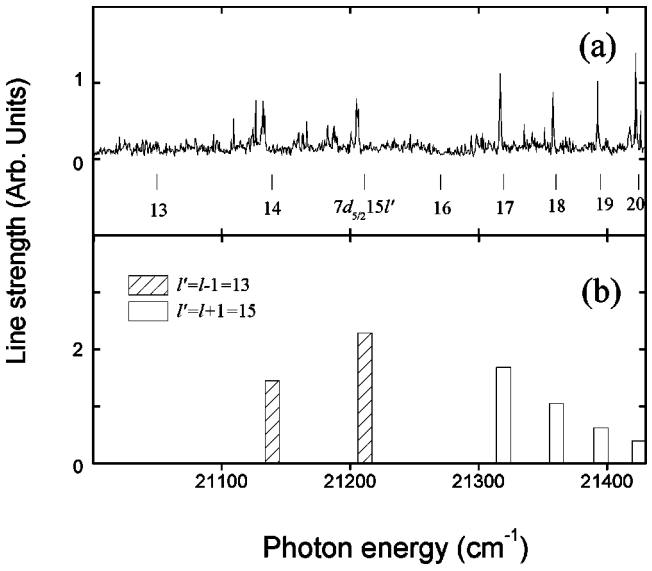


FIG. 2. (a) Excitation spectrum of $7d_{5/2}n'l'$ from $5d_{5/2}16l(l=14)$ obtained by scanning the fifth laser photon energy; (b) calculated line strengths $|D_d|^2$ for $7d_{5/2}n'l'$. The unperturbed line positions of the doubly excited states are indicated by vertical bars. The observed lines usually shift to the left as a result of the dipole perturbations (between $7dn'l'$ and $8pn''l'', 6fnl$ series). The perturbations do not influence our discussions in this paper. Note the missing $7d_{5/2}13l'$ and $7d_{5/2}16l'$ resonances as explained in the text.

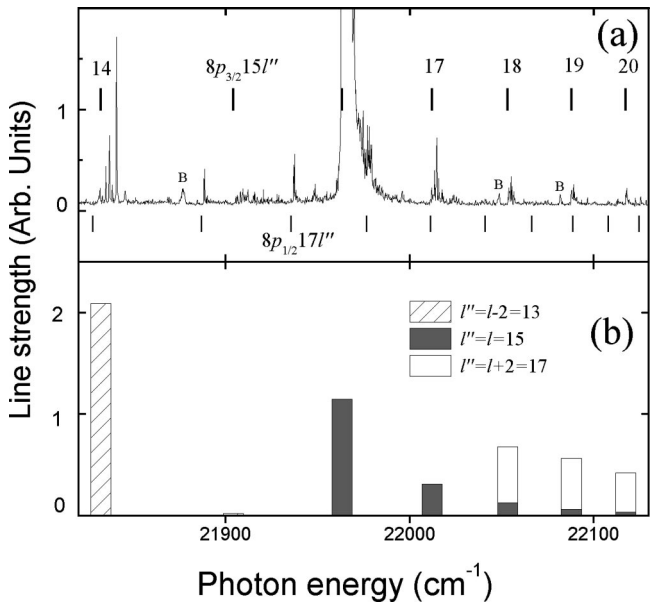


FIG. 3. (a) Excitation spectrum of $8p_{3/2}n''l''$ from $5d_{5/2}16l(l=15)$ obtained by scanning the fifth laser photon energy. Extraneous background lines are labeled by B; (b) calculated line strengths $|D_p|^2$ due to the perturbation between $6f_{7/2}16l$ and $8p_{3/2}n''l''$. Note that for $n''=16$ in (a), $|D_p|^2$ is overwhelmed by the strong resonance from the trivial process $5d_{5/2}16l \rightarrow 8p_{3/2}16l$, where the outer electron remains unchanged.

In Fig. 2(a) we show the results of a measurement where the fifth laser is scanned below the $7d$ continuum. In an ICE scheme, where correlation effects are neglected, the excitation of the $7dnl$ series is forbidden by the dipole selection rule for the transition $5d \rightarrow 7d$. The measurement, however, clearly shows a series of resonances identified as $7d_{5/2}n'l'$ states. We can assign $n$ quantum numbers to the resonances by calculating the energy positions using a simple independent-particle picture for the $7d_{5/2}n'l'$ states. The positions are indicated by vertical bars in Fig. 2(a). Two striking features are recognized: (a) below the energy corresponding to the binding energy of $7d_{5/2}n=14$ states no resonances are observed and (b) the disappearance of the $7d_{5/2}16l'$ resonance line.

In Fig. 3(a) we show a measurement where the last laser is scanned in the energy range below the $8p$ continuum. Here, the dipole excitation of the $5d_{5/2}16l(l=14,15) \rightarrow 8p_{3/2}n''l''=l$ is possible. However, the shake-up or -down process for high-$l$ states is negligible as well as the shake-off excitation (the shake-up process where the outer electron escapes) of the lower continuum channels such as $5f\epsilon l$. Consequently, the only expected excited resonance is the $8p_{3/2}n''=16l''=l$. One clearly sees the dominant excitation to the $8p_{3/2}16l''$ state. But interestingly, the excitation of other members of the Rydberg series is clearly visible, with the line strength dramatically varying.

To explain the data and to analyze the correlation, we use a perturbative configuration-interaction method. This approach has already been used to explain energy shifts for high-$l$ states [2,10,18], but has not been applied in this context to analyze line strengths, which reveal more directly the

multipole character of the electronic correlation.

Possible configurations that could act as perturber states and mix in sufficient dipole moment to excite the $7d_{5/2}n'l'$ series are the configurations $Nf_{5/2,7/2}16l$ and $Np_{3/2}16l$. As can be seen from Fig. 1 these perturber states are located relatively far away from the spectral range under study, except for the $8p_{3/2}16l$ state. However, its contribution is negligible, as will be explained later on. Our calculations show that the $7d_{5/2}n'l'$ and the $8p_{3/2}n''l''$ spectrum can be explained through the perturbation from the dipole and multipole correlation with the $6f_{7/2}16l$, respectively, which gives by far the largest contribution. It should be noted that the energy range of the $7d_{5/2}n'l'$ and $8p_{3/2}n''l''$ Rydberg series is well below the lowest bound state of the $6f_{7/2}nl$ (and also the $Nf_{7/2}nl$ with $N>6$) series. In these cases, the MQDT-type channel wave functions for these series vanish in the outer region. Thus a MQDT application including the $Nf_{7/2}nl$ channels ($N\geqslant 6$) seems to be excluded.

In the framework of CI, the nonperturbed wave functions of the high-$l$ doubly excited states can be constructed from wave functions of the $Sr^+$ ion (for the inner electron) and the hydrogenic ones (for the outer electron). The interelectronic interaction is expanded on spherical harmonics, i.e. (atomic units are used unless specified),

$$\frac{1}{r_{12}}=\sum_{k=0}\frac{r_<^k}{r_>^{k+1}}P_k(\cos\theta_{12})=\sum_{k=0}T_k. \tag{1}$$

Using perturbation theory, the perturbed wave function for the $7d_{5/2}n'l'$ state can be written as

$$\Psi_d=|7d_{5/2}n'l'\rangle+d_{n'}|6f_{7/2}16l\rangle+\cdots, \tag{2}$$

where the perturbative coefficient $d_{n'}$is

$$d_{n'}=\frac{1}{E_{n'}}\left\langle 6f_{7/2}16l\left|\frac{1}{r_{12}}\right|7d_{5/2}n'l'\right\rangle$$

$$\simeq\frac{1}{E_{n'}}\langle 6f_{7/2}16l|T_1|7d_{5/2}n'l'\rangle, \tag{3}$$

and where $E_{n'}$ is the unperturbed binding-energy difference between the two states. In Eq. (2) only the term that is important for the resonance strength is given explicitly. The dominant perturbation arises from the dipole correlation ($l'=l\pm 1$). The dipole transition amplitude can then be written as

$$D_d=d_{n'}\langle 5d_{5/2}|r|6f_{7/2}\rangle\sim d_{n'}. \tag{4}$$

Similarly for the $8p_{3/2}n''l''$ series, the dipole transition amplitude due to the perturbation of $6f_{7/2}16l$ can be written as

$$D_p=q_{n''}\langle 5d_{5/2}|r|6f_{7/2}\rangle\sim q_{n''}\quad\text{for}\quad n''\neq 16, \tag{5}$$

where

$$q_{n''}\simeq\frac{1}{E_{n''}}\Bigg(\langle 6f_{7/2}16l|T_2|8p_{3/2}n''l''\rangle$$

$$+\sum_{N',l'}\mathrm{P}\int\langle 6f_{7/2}16l|T_1|N'd_{5/2}n'l'\rangle_{\mathrm{E}}$$

$$\times\frac{{}_{\mathrm{E}}\langle N'd_{5/2}n'l'|T_1|8p_{3/2}n''l''\rangle}{E_{n''}-E_{n'}}dE_{n'}\Bigg). \tag{6}$$

Here the subscript ''E'' denotes the wave function as energy-normalized; the integral includes both discrete and continuum states. The perturbation includes the quadrupole and the dipole-dipole cross-term correlations. These two terms are of the same order and constructive for most of the observed resonances. It is the two dipole moments of Eqs. (4) and (5) that lead to spectral structures beyond what is expected from the simple ICE scheme.

Our theoretical simulation for Fig. 2(a) is based on Eq. (4). The calculated results are shown in Fig. 2(b). The comparison with the experimental line strength shows that the vanishing states are correctly described by our analysis. Furthermore, the calculations allow the identification of the angular momentum of the excited $7dn'l'$ series, which is $l'=l+1$ for the states with $n'>16$ and $l'=l-1$ for states with $n'<16$. The vanishing excitation of the $7d_{5/2}16l'$ state is a direct consequence of the selection rule of the matrix element $\langle 16l|1/r^2|16l'\rangle$, which is a part of the radial integral in Eq. (3). It vanishes exactly for hydrogenic wave functions with $l'=l\pm 1$ [19]. The perturbation from $6f_{5/2}16l$ is neglected in Eqs. (2)–(6) because the dipole transition from $5d_{5/2}$ to $6f_{7/2}$ is much stronger than to $6f_{5/2}$ as a result of elementary angular-momentum coupling considerations.

Qualitatively, there is still some discrepancy concerning the relative line strength. This stems partly from the detection process, which favors the detection of the higher $n$-state members of the Rydberg series [20,21]. Considering this experimental fact our calculated result is very consistent with the experimental data shown in Fig. 2(a).

It is noted that using the $8p_{3/2}16l$ as a perturber state would lead qualitatively to similar results. However, the dipole transition matrix element in Eq. (4) for the $5d\rightarrow 8p$ transition is more than two orders of magnitude smaller than the $5d\rightarrow 6f$ transition, so that the $8p_{3/2}16l$ perturber state can be safely excluded [22].

To explain the $8p_{3/2}n''l''$ resonances, we have performed a similar calculation based on Eq. (5). In Fig. 3 we show the experimental and calculated spectrum of $8p_{3/2}n''l''$ for $l=15$. Figure 3(b) shows that the maximum of CI between $8p_{3/2}n''l''$ and $6f_{7/2}16l$ takes place in different energy regions for different $l''$, i.e., at $n''<16$, for $l''=l-2$, at $n''>16$ for $l''=l+2$, and around $n''=16$ for $l''=l$. The calculated line strength at $n''=16$ due to the perturbation of $6f_{7/2}16l$ is overwhelmed by the strong resonance from the trivial process $5d_{5/2}16l\rightarrow 8p_{3/2}16l$, where the outer electron keeps unchanged. There are several striking features in the spectrum in Fig. 3(a) that are confirmed by the calculation. It shows that the correlation effects are indeed responsible for the satellite structure.

For $n'' \neq 16$, the calculated line strengths reproduce well the observed ones, especially the vanishing line strength at $n''=15$. Furthermore, nicely reproduced is the significantly enhanced resonance $8p_{3/2}n''l''(n''=14)$, which is not expected in the shake process due to $n''<l+1$. For $n''>l+1=16$ resonances, the spectrum looks like a shake spectrum, but the calculation presented in Fig. 3(b) shows that it is dominated by $l''=l+2$ for $n'' \geqslant 18$ other than $l''=l$ in the simple ICE process. This implies that it may be necessary to modify the theoretical approach for the shake-off excitation in which the angular momentum of the outer electron is conserved [2].

In the above discussion, we have ignored the perturbation between $8p_{3/2}16l$ and $8p_{3/2}n''l''(n'' \neq 16)$. The contribution of the $8p_{3/2}16l$ perturber state to $D_p$ is similar to Fig. 2(b) but much less for larger $|n''-16|$. Expressions involving this part need the accurate value for $\langle 5d_{5/2}|r|8p_{3/2}\rangle$ by calculation or fitting experimental data, which will not be discussed here. In any case, it influences only the line strengths of $8p_{3/2}n''l''$, with $n''=16\pm1$ slightly due to the small value of $|\langle 5d_{5/2}|r|8p_{3/2}\rangle|$ $(\sim 0.01)$. For the same reason, as already mentioned, we have ignored the contribution from the perturbation of $8p_{3/2}16l$ in Fig. 2(b). Some resonant lines of $8p_{1/2}n''l''(n''=16-18,19)$ in Fig. 3(a) are also partly due to the perturbation from $8p_{3/2}16l$, for which additional influences are from the correlation effects in the initial state $5d_{5/2}16l$.

Finally, we mention that the specific electron-electron interaction can be understood semiclassically. The inner turning point $P_{\text{in}}$ of the outer electron orbit plays the central role for the correlation. The maxima for the CI discussed above will occur when two configurations have the same $P_{\text{in}}$. Thus for $l'$ (or $l''$)$>l$, the maxima occur at $n'$ (or $n''$)$>n$ and vice versa, as shown in the above calculations. A similar spectrum should be observable for higher-pole correlations (such as, e.g., the octupole). Because these types of spectra are dominated by the inner region, they provide very sensitive ways to verify the theoretical calculations involving electronic correlation.

In conclusion, we have observed characteristic features in the resonant multiphoton excitation of high-angular-momentum states in Sr. The spectra reveal the importance of the multipole electronic correlations in the excitation process. Most striking is the direct observation of a selection rule for the dipole correlation manifested in a vanishing line strength. The excitation scheme provides an alternative and very detailed way to study the electronic correlation in the high-$l$ asymmetric states directly. It should be pointed out that there is no penetration between the two electrons in the asymmetric states. This is different from the case of ''intrashell'' states in light atoms. The experiments also provide possible ways to realize very-high-$l$ states beyond the limit of the Stark-switching technique through multiphoton up-and-down excitation.

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