Dipole Structure of Planetary Atoms

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Using resonant multiphoton laser excitation in combination with temporarily applied electric fields we have excited high-lying planetary states in strontium. Thereby, the angular momenta were high enough to prevent penetration of the Sr^{++} core, as well as the mutual penetration of the two excited wave functions. Analysis of the spectra (quantum defects and oscillator strength modulations) clearly revealed the dipole structure of such a "planetary atom," where the outer electron is bound to either a permanent or an induced dipole formed by the inner electron and the ionic core.

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It has long been realized that the atomic three-body Coulomb system (two highly excited electrons and one positive ion) can be divided into two conceptually different classes of states: the "intrashell states" (among them the famous Wannier states) where the electrons move at roughly equal distances $\langle r_1\rangle$ and $\langle r_2\rangle$ from the ion, and the "intershell" or "planetary states" where the electrons are predominantly located at different radial distances $\langle r_1\rangle \neq \langle r_2\rangle$, having different quantum numbers n_1 and n_2. Both classes have long been subject to vigorous theoretical investigations, and lower-lying members have already been investigated in some detail. This is not true for high-lying states where such fundamental properties as quantal classification schemes [1–4] or the stability of states [5] are still under dispute. The cause for this ongoing discussion—and perhaps its most remarkable aspect—is the lack of comprehensive experimental surveys on high-lying atomic three-body Coulomb states. State-of-the-art vacuum ultraviolet (VUV) photoexcitation experiments in H^- [6] and He [7] are limited in resolution to doubly excited states n_1n_2 with $n_1 \sim 7$ and $n_2 \sim 16$. More seriously, they have revealed a quasiselection rule which limits one-photon excitation from the ground state almost exclusively to states with one specific set of pair quantum numbers [8,9]. This is a rather unfortunate restriction as it leaves the majority of three-body Coulomb states inaccessible for one-photon experiments, regardless of any further improvement in the resolution.

Alternatively, three-body Coulomb states can be investigated by multiphoton laser excitation, using alkaline-earth atoms [10–13]. In principle, this method has the potential of overcoming the above limitations both in the resolution *and* in the one-photon selection rules. The essential task when using heavy atoms is to minimize core penetration, which spoils the characteristic SO(4) Coulomb symmetry [3] of the two-electron–ion subsystems. Core penetration can be avoided by excitation of both electrons into sufficiently high angular momentum states l, whereby $l \geq 3$ and $l \geq 4$ suffices even for the heaviest alkaline-earth atoms Sr and Ba, respectively. Various experiments exist where either the outer or the inner of two excited electrons is non-core-penetrating, and effects of long-range correlations have been observed in the latter case [11]. However, none of these studies could reveal the full spectrum of a planetary atom, as the outer electron was either dominantly in an l eigenstate or so highly excited that its effect was that of an unspecified "frozen" charge distribution near the classical outer turning point.

In the present Letter we report on experiments where these conceptual restrictions have finally been removed. Using a further extension of the multiphoton laser excitation we were able to start from outer electron Rydberg states $n_2'l_2'$ with well-defined high angular momentum $9 \leq l_2' \leq n_2' - 1$, which was not only high enough to prevent penetration of the Sr^{++} core, but even high enough to prevent penetration of the inner electron's final-state wave functions n_1l_1 with $l_1 \geq 3$ [in terms of classical turning points a sufficient condition is $l_2(l_2+1)/2 > 2n_1^2/Z_1$, where $Z_1 = 2$ is the core charge seen by the inner electron]. Thereby, n_2 was now kept sufficiently low to resolve spectral structures of the outer electron.

The apparatus and the resonant excitation method for the inner electron is the same as described earlier [11]. The new feature introduced in the present experiment was the excitation of the outer electron in the presence of a static electric field ($F \sim 300$–1000 V/cm), exciting one specific Stark level n_2k out of the nondegenerate Stark manifold. Adiabatic reduction of the field transforms the Stark state into one specific angular momentum eigenstate $n_2'l_2'$ ("Stark switching technique" [14,15]). After the field had returned to zero (actually slowly crossing through zero) the second valence electron was excited by the usual resonant multiphoton scheme via the intermediate autoionizing resonances $5pn'l_2'$ and $5dn'l_2'$ allowing achievement of non-core-penetrating states with $l_1 \geq 3$. The final state may be designated as (n_1,l_1,n_2,l_2); alternatively, for strongly correlated states we will use the notation (n_1,n_2,γ), where γ stands for any set of suitable correlation quantum numbers. The wavelength of the last laser was scanned over a broad range from 340 to 580 nm, covering principal quantum numbers n_1 of the inner electron in the range 5 to 8. Detection of doubly excited states proceeded again through autoionization and subsequent photoionization of the fragments, whereby Sr^{++} ions were finally detected in a pulsed time-of-flight mass analyzer.

Before going into the discussion of the experimental results it is useful to recall the physics of planetary states.

Usually, they are discussed in the dipole approximation (for recent reviews see Refs. [3] and [16]), where the outer electron moves in the radial potential

$$V(r_2) = -1/r_2 + A/r_2^2 , \quad (1)$$

with the "dipole channel operator" A [3] being given by

$$A = l_2^2 - n_1(\mathbf{b}_1 \cdot \hat{\mathbf{r}}_2) . \quad (2)$$

Here, $\mathbf{b}_1$ is the Runge-Lenz vector of the inner electron, which is a constant of motion if the SO(4) symmetry of the inner ion is preserved (i.e., if all ionic angular momentum states are degenerate). Apart from this, the only essential condition is the relation $n_2 > n_1$, which implies that the classical orbiting time τ_2 of the outer electron is large compared to the orbiting time τ_1 of the inner one (adiabaticity condition). Under these circumstances the correlated wave functions of the inner electron are essentially the same as the Stark wave functions discussed previously in the "frozen planet approximation" [11]. According to the possible relative orientations of $\mathbf{b}_1$ and $\hat{\mathbf{r}}_2$ the potential (1) leads to a number of nondegenerate Rydberg series of outer electron states n_2, all converging to the same excited state n_1 of the ion.

Next, we consider the situation when the angular momentum splitting of the inner electron can no longer be neglected, as is the case in doubly excited alkaline-earth atoms, in doubly excited He^-, and even in doubly excited helium, if relativistic effects are taken into account. It is instructive to discuss this in terms of a precession of the Runge-Lenz vector $\mathbf{b}_1$, with its orbiting time τ_{RL} introducing the third characteristic time scale of the system. τ_{RL} is inversely proportional to the change in the inner electron's quantum defect with l_1 at the energy E, written as [17]

$$\tau_{RL}(E) = \tau_1 \left(\frac{\partial \mu_{l_1}(E)}{\partial l_1} \right)^{-1} . \quad (3)$$

Now the adiabaticity condition must additionally ensure that the outer electron can follow the precession of the Runge-Lenz vector, i.e., $\tau_{RL} \gg \tau_2 \gg \tau_1$. Using $\tau_i \propto n_i^3/Z_i^2$ (where Z_i are the effective core charges of electrons $i = 1,2$) this can be rewritten as

$$\left(\frac{\partial \mu_{l_1}}{\partial l_1} \right)^{-1} \gg 4 \left(\frac{n_2}{n_1} \right)^3 \gg 1 . \quad (4)$$

This modified adiabaticity condition may only be met for not too large quantum defects μ_{l_1} and not too high outer quantum numbers n_2; however, in any series of dipole states (n_1, $n_2 \to \infty$, γ) it will be violated as the outer electron will eventually become decoupled from the precession of the Runge-Lenz vector. Note that this situation is very similar to angular momentum recoupling in molecular Rydberg states [18], discussed in terms of frame transformations in quantum-defect theory (QDT). In planetary atoms the long-range correlation forces prevent the application of Coulomb-type QDT; instead, one obtains at high n_2 a new effective potential for the outer electron of the form

$$V(r_2) = -\frac{1}{r_2} + \frac{\tilde{l}_2(\tilde{l}_2+1)}{2r_2^2} + \frac{\tilde{\alpha}}{r_2^4} , \quad (5)$$

where $\tilde{l}_2$ and $\tilde{\alpha}$ are channel parameters of the coupled system [19] with the asymptotic behavior $\tilde{l}_2 \to l_2$ and $\tilde{\alpha} \to \alpha$ for $n_2 \to \infty$. Here α is the dynamic polarizability of the inner electron's wave function in the angular momentum eigenstate $n_1 l_1$; it is defined for all l_1 (including core penetrating states), scales as n_1^7, and can be calculated numerically. A potential such as (5) leads to a single Rydberg series of states with a nonzero quantum defect μ_2 which, for nonoverlapping wave functions (as in the present experiments), is entirely determined by $\tilde{\alpha}$ and $\tilde{l}_2$.

Figure 1 shows a typical example of a planetary spectrum, divided into regions "a," where n_2 is sufficiently high that (4) is clearly violated and (5) is expected to hold, and "b" at lower n_2, together with an additional structure "c" to be discussed later. The series of states in region a, with $n_2 \geq 20$, converges to the $6g$ limit of Sr^+, and is excited out of the ($5d_{5/2}$, $n_2' = 17$, $l_2' = 9$) intermediate state. Note that this excitation obviously violates the independent-particle selection rules, which alone is a sign of strong electron correlation in the final states. Figure 2 shows, in a separate and more detailed scan, the transition from region a to region b. Analysis of the isolated Rydberg levels in region a reveals a constant quantum defect $\mu_2(\text{mod}1) = 0.79$. Such a large quantum defect would be totally surprising for a series of high l_2 Coulomb states, but is consistent with dipole states of the potential (5). In fact, μ_2 itself (which could not be determined in the present experiment) is likely to be even

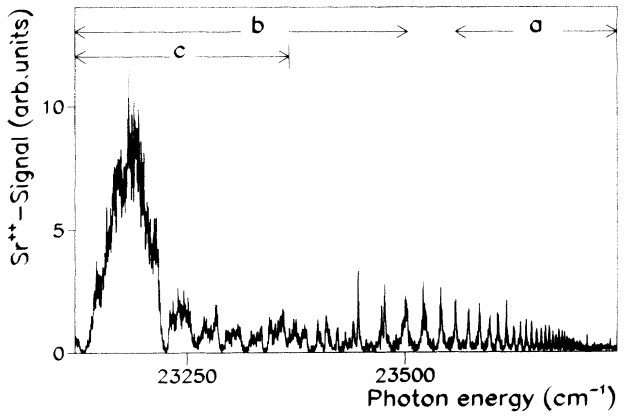

FIG. 1. Typical planetary spectrum, observed after excitation of the intermediate $5d_{5/2}$, $n_2' = 17$, $l_2' = 9$ state, as a function of photon energy of the last laser. The features occurring in the energy regions "a," "b," and "c" are separately discussed in the text.

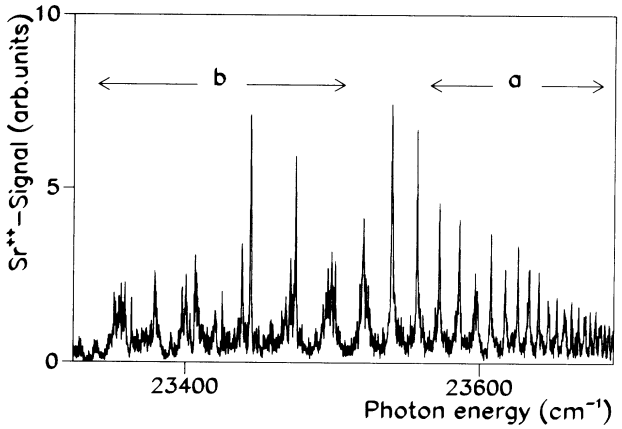

FIG. 2. High-resolution scan of the transition from region "a" [asymptotic Rydberg series ($6g$, $n_2 \rightarrow \infty$, l_2), belonging to the potential (5)] to region "b" at lower n_2, showing the evolution of different series (n_1,n_2,γ).

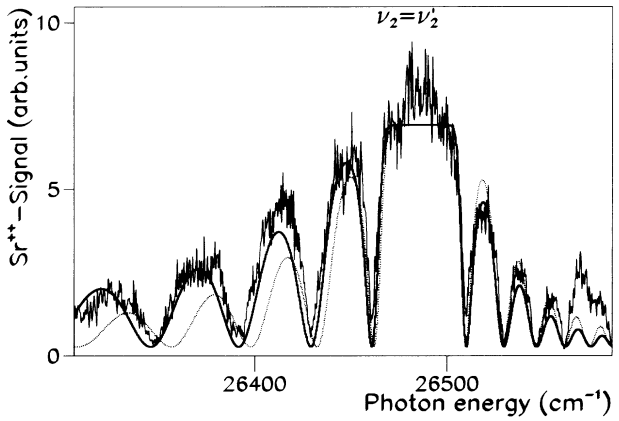

FIG. 3. Detailed study of an observed and calculated (solid line) overlap structure $|\langle\psi^P(\nu_2)|\psi^C(\nu_2'=21)\rangle|^2$, similar to feature "$c$" in Fig. 1, but taken at higher laser power and showing some saturation effects [21]. The final planetary state $\psi^P(\nu_2)$ is calculated in the potential (5), using the independently calculated polarizability $\alpha=5\times10^5$ a.u. of the core state (here Sr^+ $7f$). Note the characteristic phase shift and height asymmetry of the side maxima when compared with a pure Coulomb state $\alpha=0$ (dotted line).

much larger: Using the asymptotic polarizability $\alpha=5.25\times10^5$ a.u. of the Sr^+ $6g$ state we calculate $\mu_2=6.68$, which is in good (perhaps fortuitous) agreement with the observed $\mu_2(\mathrm{mod}1)$. Various asymptotic planetary Rydberg series (n_1, l_1, $n_2\rightarrow\infty$, l_2) have been analyzed this way, exhibiting rather large quantum defects throughout, which are tabulated in Table I.

For decreasing n_2, Fig. 2 exhibits the expected deviations from a single Rydberg series in region b, showing an apparent splitting of the individual lines into various sublevels. For further decreasing n_2 these eventually overlap to form a dense spectrum of rather narrow levels with no obvious regularity. This tendency was observed in all planetary Rydberg series, and is again consistent with the above discussion of the general structure of planetary spectra: In region b we gradually enter the region where relation (4) holds. As a result, the different series (n_1,l_1,n_2,l_2) will rearrange themselves to form new series (n_1,n_2,γ), according to the spectrum of the dipole channel operator A. However, we do not necessarily expect the evolution of the pure dipole channels since around $n_1<10$ a large fraction ($\sim30\%$) of the Sr^+ angular momentum states still have quantum defects of the order of unity [20]. A detailed investigation of this transition region has not been attempted, as it requires the solution of the three-body Hamiltonian in the region between the limiting cases (1) and (5).

TABLE I. Quantum defects $\mu_2(\mathrm{mod}1)$ and series limits for different planetary series in region a ($n_2\gtrsim20$) obtained by a fit of the data by a one-channel Rydberg formula. The line positions followed the fit formula to within the laser resolution (0.2 cm^{-1}). The sign of the quantum defects has been determined from calculations, using the asymptotic value α for the polarizability of the Sr^+ n_1l_1 core. The series limits are given with respect to the ionic ground state Sr^+ $5s$.

Transition	$\mu_2(\mathrm{mod}1)$	Series limit (cm^{-1})
$5d_{5/2}$; $n_2'=17$, $l_2'=9\rightarrow 5g$; n_2l_2	−0.03	71353.9
$5d_{5/2}$; $n_2'=17$, $l_2'=9\rightarrow 6g$; n_2l_2	0.79	76731.6
$5d_{5/2}$; $n_2'=17$, $l_2'=11\rightarrow 7h$; n_2l_2	−0.09	80000.4
$5d_{5/2}$; $n_2'=16$, $l_2'=10\rightarrow 8p$; n_2l_2	−0.21	79126.2
$5d_{5/2}$; $n_2'=16$, $l_2'=10\rightarrow 8d$; n_2l_2	0.53	78702.2

As shown by feature c in Fig. 1, a localized periodic modulation of the density of states was found in some cases, embedded in region b of the spectrum. It resembles very much the well-known "overlap structure" which arises in regular Rydberg atoms when a discrete ionic core transition is excited with variable laser frequency [21]. There, the energy mismatch between the core transition and the laser frequency is compensated by the Rydberg electron, which undergoes a shake (or monopole) transition from its initial Coulomb state $\nu_2'l_2'$ to a final Coulomb state ν_2l_2, where ν is the effective quantum number. The squared monopole matrix element $|\langle\nu_2|\nu_2'\rangle|^2$ introduces a characteristic energy dependence in the cross section [21], which is independent of channel mixing and resonance structures in the final states, and only probes the Rydberg character of the outer electron.

As we expect structure c, located in the region $\nu_2\sim\nu_2'$, to be a similar cross-section modulation we have calculated the energy dependence of the squared overlap integral between an initial Coulomb state $\psi^C(\nu_2',l_2')$ and a final planetary state $\psi^P(\nu_2,l_2)$ which is an eigenstate of the Hamiltonian with dipole potential (1) or (5). The result (shown in detail in Fig. 3) is highly interesting, showing that the squared overlap integral $|\langle\psi^P(\nu_2)|\psi^C(\nu_2')\rangle|^2$ is a sensitive indicator of the non-Coulombic properties of final planetary states. These properties induce a notice-

able shift in the zeros of the overlap structure, and a left-right asymmetry in the height of the side maxima, which depend on both the sign and magnitude of the core polarizability $\tilde{a}$ [or, similarly, on the dipole operator A in Eq. (1)]. While a possible shift of the zeros was already discussed by Boulmer *et al.* [22] [considering the energy eigenvalues of (5) in the basis of pure Coulomb waves] we point out that the full calculation of the overlap integral with dipole waves reproduces not only the observed shift over a large energy interval but also the asymmetry of the overlap structure which was observed here for the first time. Hence, we are left with the interesting observation that—although we cannot identify any individual resonances in region b, where the density of states is almost uniform—we have still unambiguous information about the global dipole structure of these states, which is exploited through their projection on the well-defined initial Coulomb state $\psi^C(\nu_2',l_2')$ via the overlap integral.

In conclusion, we have performed the first laser investigation of the internal dipole structure of planetary atomic states (n_1,n_2,γ) with $n_1 < n_2$, where both the penetration of the core *and* the mutual penetration of the two excited wave functions was suppressed. In resolving planetary states as high as $n_2 \sim 60$ we were able to observe the transition from *induced* dipole states of the inner electron, which prevail in the limit $n_2 \to \infty$, towards *Coulombic* (permanent) dipole states, where the outer electron becomes strongly coupled to the inner Runge-Lenz vector. Furthermore, we were able to measure and calculate the energy dependence of a shake transition matrix element of the outer electron, which yields quantitative information on the planetary dipole structure even when individual resonances of the system cannot be resolved.

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