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Metastable Autoionizing States in Sodiumlike Chlorine*

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We have observed metastable autoionizing states associated with partially stripped, highly excited chlorine ions. Charge-state-fraction, electron-energetic, and series-limit arguments indicate that these states are primarily associated with *sodiumlike chlorine* (Cl^{6+}), but few energy-level calculations exist for firm identification. Prominent peaks occur in the electron spectrum at 90 ± 3, 101 ± 3, 138 ± 3, and 182 ± 3 eV. A long-lived component of the 182-eV peak has a lifetime ⩾ 43 nsec.

We report here experimental evidence for the existence of significant numbers of highly metastable autoionizing states associated with partially stripped, highly excited chlorine ions. Figure 1 shows the spectra of energy-analyzed electrons that were emitted in autoionizing decays of these states in the energy range ~70–220 eV (in the rest frame of the emitting ion). We have recently reported[1] on the study of a metastable autoionizing state ($1s2s2p\,^4P_{5/2}$) associated with lithiumlike chlorine (Cl^{14+}) and argon (Ar^{15+}) ions. The autoionizing electrons emitted by those highly stripped ions were of considerably higher energy (~2000 eV) than in the present experiment in which ions in much lower charge states are of importance. We believe that the observed spectral features are primarily due to the decay of metastable autoionizing states associated with sodiumlike chlorine (Cl^{6+}). While some evidence for the existence of such states in the neutral alkali atoms has been shown by Feldman and Novick,[2] to our knowledge the present work is the first study of a sodiumlike system in which energy analysis of the emitted electrons has been exploited. Firm identification of the electron spectral lines is at present precluded by an almost complete lack of theoretical calculations of the energies and lifetimes of such states. There

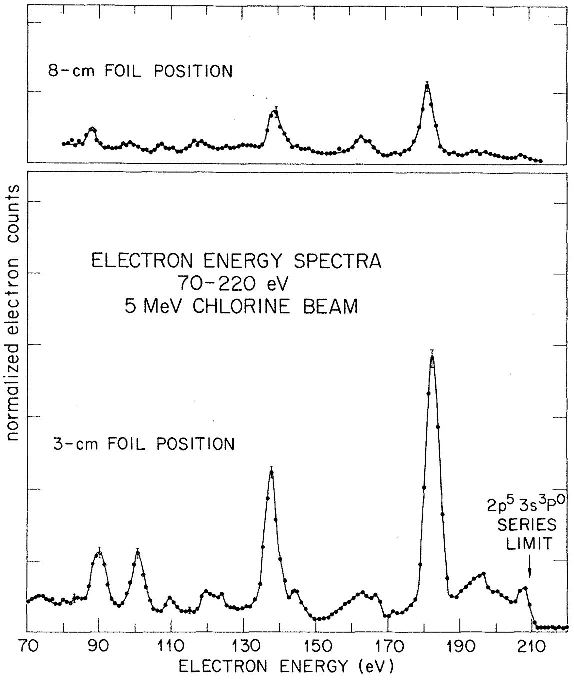

FIG. 1. Segment of electron spectra from 5-MeV chlorine ions undergoing decay in flight, plotted in the ionic rest frame. Data are shown for two different target positions, 3 and 8 cm from the spectrometer viewing region.

is now clearly a need for such calculations.

Our electron spectroscopic technique has been described in detail previously.[3] Briefly, an ion beam from the Oak Ridge tandem accelerator was passed through a thin carbon foil (~15 $\mu g/cm^2$) which served both to strip and excite the ions of the beam. Electrons emitted by spontaneously decaying ions in the emergent beam were energy analyzed by a cylindrical mirror analyzer and could be counted as a function of separation of the foil target from the spectrometer viewing region. In the present experiment chlorine ions of ~5 MeV energy were incident on the foil target. This energy was chosen to maximize the production of sodiumlike chlorine (Cl^{6+}) ions in the beam for the specific purpose of searching for especially metastable systems of alkalilike structure. At this energy ~30% of the emergent ion beam consists of Cl^{6+} ions.[4] Significant fractions of ions in adjacent charge states are also present, but for reasons discussed below it is unlikely that these ions contribute appreciably to the observed spectrum unless certain multiple-excitation conditions are present.

It is plausible that many of the states that are observed in decay are quartet states of the ion Cl^{6+} that are formed from core-excited configurations such as $2p^5(nl)(n'l')$ or $2s2p^6(nl)(n'l')$ with $n, n' \geq 3$, in which a single electron has been excited from a previously closed shell or subshell into an outer orbital. It is also conceivable that the high metastability of the states that we observe indicates the population of states of even higher multiplicity, arising from the simultaneous excitation of two or more inner-shell electrons. These multiple core-excited configurations might also occur for the ions in the beam in adjacent charge states (fluorine-, neon-, magnesium-, or aluminum-like chlorine), which might account for some of the observed spectral features. However, the existence of such metastable autoionizing states in these ions or members of their isoelectronic sequences have never, to our knowledge, previously been observed. While the possibility of multiple core excitations may not be ruled out it is clear from simple energetic arguments that autoionizing electrons emitted from single core-excited ions in adjacent charge states would lie outside of the energy range that has been studied in this experiment. Moreover, we observe in the spectral data the series limit that would be expected for single-excited sodiumlike states.

The energies of the electrons shown in Fig. 1 are given in the rest frame of the emitting ion. These electron energies are the same as the energies of the metastable autoionizing states relative to the ground state of the ion of the next highest stage of ionization, i.e., the final state of the residual ion after the autoionization process has occurred. Thus to find the excitation energy of the state one must add the first ionization potential of the relevant ion to the electron energies that are shown in the spectra of Fig. 1. Metastable autoionizing states associated with an ion in a particular charge state (and describable as single-electron core excitations) can in general have energies in the range between the ground state and the first excited state of the ion of the next highest stage of ionization, since levels higher than this limit can autoionize via the Coulomb interaction. Thus the series limit for the ion Cl^{6+} will be the excitation energy of the $2p^5 3s\,^3P^0$ state of Cl^{7+}. This energy is tabulated[5] as 209.46 eV (relative to the Cl^{7+} ground state) and agrees very well with the rather well-defined dropoff in intensity of our spectral data at the same energy (see Fig. 1). The series limits for metastable autoionizing states (describable in terms of single-electron core excitation) associated with all the ions in adjacent charge states are well below the energy range of the electrons observed. This precludes the possiblity of spectra contamination from such states that might be associated with these ions.

The absolute energies of the spectral lines in Fig. 1 are subject to certain systematic errors. The chief contributions to these errors stem from uncertainties in the foil thickness, and from small variations in the angle of admittance to the electron spectrometer with steering of the ion beam. Foil thickness uncertainty implies an uncertainty in the energy loss of the ions in the foil and hence in both the energy and energy spread of the emergent beam. These systematic uncertainties contribute to kinematic peak shifts when laboratory energies are transformed to those of the emitting ions' rest frame. By comparison, the elastic scattering of electrons showed that errors in the spectrometer calibration were negligible. The estimated energy loss in the foil contributes a measurable shift to each peak energy; all data have been compensated accordingly. Energy straggling in the foil spreads the peak energies and thus decreases the spectral resolution. This effect can be observed if one compares the spectral resolution (~2%) in the present data (for a ~5-MeV chlorine beam) with that (~0.6%) obtained in our previous work on lithiumlike chlorine[1] where a 42-MeV beam was employed. The straggling effect could clearly become a serious limitation to the method for heavy ions at much lower beam energies.

The energies of the four most prominent spectral features in Fig. 1 are 90 ± 3, 101 ± 3, 138 ± 3, and 182 ± 3 eV, where the error limits reflect the aforementioned systematic uncertainties. The energies of the spectral features relative to the assumed series limit are more accurately measurable because of the rather well-defined dropoff in intensity in the spectrum at this energy value. The peak at 101 ± 3 eV agrees well with a recent unpublished calculation by Weiss[6] of the energies of the ^4S, ^4P, and ^4D states which are all formed from the core-excited configuration $2p^53s3p$ of sodiumlike chlorine. Weiss also estimates that the energy of the $2s2p^63s3p\ ^4P$ state in Cl^{6+} is approximately 170 eV, which is close to a less prominent spectral feature in Fig. 1. An estimate of the energies of core-excited configurations such as $2p^53sns$ ($n \geq 4$), $2p^53snp$ ($n \geq 3$), and $2p^53p^2$ can be made by using the simple screening rule that the energy of a state of an ion formed by single-electron core excitation is approximately equal in energy to that of the similar state (with no core excitation) of the ion of the same charge state but of an atomic number that is increased by unity (after the appropriate ionization potential has been added). For example, the energy of the $2p^53s3p$ configuration in Cl^{6+} is approximately equal to that of the configuration $2p^63s3p$ in Ar^{6+} (after addition of the ionization potential of Ar^{5+}). While the accuracy of the simple screening method is not sufficient to identify positively the observed lines, the results of such approximate calculations do serve to show that several of the spectral features could be accounted for by the aforementioned configurations of Cl^{6+}.

Spectra were taken at selected target positions over a range of approximately 25 cm (~ 50 nsec) in order to study the decay characteristics of the most prominent peaks of the spectrum. A comparison of two such spectra taken at 3 cm and 8 cm downstream from the target is shown in Fig. 1. A similar spectrum taken at about 25 cm from the target showed that the four most prominent features (as well as traces of a few others) were still present, indicating long lifetimes for the metastable autoionizing states associated with them. A more detailed study was made of the decay in flight of the 182 ± 3-eV peak by the standard method of measuring the counting rate (per unit beam current) as a function of target position over a range of approximately 25 cm. The resulting decay curve was found to be complex in the sense that more than one exponential was needed to fit the data. It was obvious, however, that the decay curve had a very long-lived component associated with it, the decay rate of which was difficult to measure accurately because of a scatter in data points at large distances from the target. This effect may have been due to divergence of the beam after multiple scattering in the foil. Rather than quote a lifetime based upon such measurements, we choose instead to give a lower limit for the lifetime of the long-lived component associated with the 182 ± 3-eV peak. This limit is 43 nsec. To explain the high metastability of these states, one may postulate the population of a level of high multiplicity and maximum J value which cannot directly autoionize via the Coulomb interaction or mix with close-lying, fast-decaying autoionizing levels of a different multiplicity. Such a level would presumably autoionize via the weaker spin-spin interaction. This level must also be metastable against radiative decay, since the lifetime of the level is determined by the total transition probabilities for both autoionizing and radiative decay processes. It is possible to account for radiative metastability in several ways. The lowest-lying level of given multiplicity is metastable because of the selection rule on spin. The absence of a lower-lying hole of appropriate angular momentum and parity is a second possibility. While small radial transition moments sometimes occur, it seems more likely that the angular-momentum and parity selection rules are primarily responsible.

The existence of many long-lived and highly excited states in alkalilike systems has been clearly shown by this work and our previous investigations. These states could conceivably play an important role in physical processes in which energy storage and secondary ionization are of consideration.

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Observation of Higher-Order Acoustic Paramagnetic-Resonance Transitions in MgO:Fe^{2+}†

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The higher-order $\Delta m = 3$ and $\Delta m = 4$ transitions in MgO:Fe^{2+} (effective spin $= 1$) have been observed directly by acoustic paramagnetic resonance. The angular dependence and temperature dependence are found to differ significantly from the normally "allowed" $\Delta m = 1$ and $\Delta m = 2$ transitions. A theory due to Fedders and Myles agrees well with the observed behavior of the $\Delta m = 4$ line.

We report, in this Letter, the observation of higher-order acoustic paramagnetic-resonance (APR) transitions in MgO:Fe^{2+}. The resonance lines occur, for a given frequency, at magnetic field values corresponding to $\Delta m = 3$ and $\Delta m = 4$ transitions, normally forbidden for APR. Since Fe^{2+} in MgO constitutes an effective spin-1 system, $\Delta m = 3$ and $\Delta m = 4$ transitions cannot be explained on the usual basis of transitions induced among single-ion energy levels.[1] The intensities and angular dependencies (i.e., dependence on angle θ between the externally applied magnetic field $\vec{H}$ and the direction of propagation $\hat{q}$ of the longitudinal acoustic waves) of the higher-order lines differ significantly, also, from those observed for the APR due to exchange-coupled pairs of ions, first reported by Guermeur *et al.*[2] for MgO:Ni^{2+} and also observed by us in the present specimen of MgO:Fe^{2+}. The present results agree well, on the other hand, with a virtual-spin-state mechanism described by Fedders and Myles in the following Letter.[3]

The present measurements were made on a single crystal of MgO, cleaved along (100) planes and prepared with end faces optically flat and parallel. The crystal was the same (MgO No. 1) as that used by Leisure and Bolef[4] in their study as a function of temperature of the Fe^{2+} $\Delta m = 2$ transition. The concentration of Fe^{2+} ions was measured by electron paramagnetic resonance at 10 GHz to be approximately fifty parts per million. Longitudinal acoustic waves were generated and detected by use of vacuum-deposited thin-film CdS piezoelectric transducers. The sensitive continuous-wave transmission APR spectrometer has been described previously.[5] Measurements were made over a temperature range of 4.2 to 15 K and at frequencies from 1.1 to 1.3 GHz.

While slowly scanning the magnetic field near 100 Oe, two absorption lines were observed in addition to the expected intense APR line at $g = 6.8$ ($\Delta m = 2$ transition). In Fig. 1 are shown the

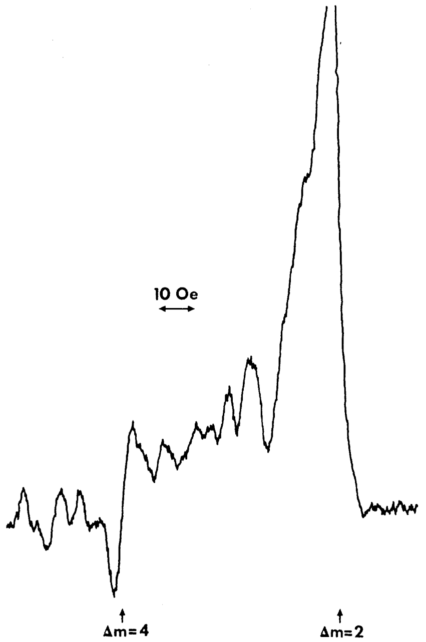

FIG. 1. Recorder trace of $\Delta m = 2$ and 4 APR lines (first-derivative traces) at $\theta = 90°$ for longitudinal acoustic waves of frequency $\nu = 1.108$ GHz propagated along the [100] axis of MgO:Fe^{2+} No. 1. $T = 4.2$ K.