Giant-Dipole-Resonance Absorption in Atomic Thorium by a Novel Two-Laser Technique

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A novel two-laser technique for photoabsorption studies of refractory and corrosive elements is described and applied to observe for the first time the absorption spectrum of atomic thorium. Results on the giant $5d$ dipole resonance in the extreme uv are found to be in good qualitative agreement with recent theoretical predictions. The spectrum is very similar to that reported for the metal, indicating that the $5f$ wave function is largely unaffected by its solid environment.

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Photoabsorption studies of atomic systems can provide fundamental information, particulary about inner-shell and double-electron transitions that cannot be obtained by more direct emission observations. Such transitions usually lie in the vacuum ultraviolet or extreme uv region of the spectrum. Even at longer wavelengths, absorption spectra, originating as they do from one or a limited number of low-lying levels, are valuable in unraveling the term systems of more complex atoms. Despite this interest the absorption spectra of a number of important elements (e.g., tantalum, tungsten, thorium) have not yet been observed in the vapor phase. These somewhat surprising gaps in our knowledge arise because the elements concerned are highly refractory and/or corrosive so that conventional techniques for generating an enclosed vapor are not applicable. In the present Letter we describe a novel technique for obtaining the absorption spectra of very refractory elements, even if their vapors are corrosive, and we illustrate the method by reporting for the first time the absorption spectrum of atomic thorium.

The technique is based on the dual-plasma system that has been used successfully for the controlled study of ionic absorption.[1,2] In our initial experiment[1] the output beam of a Q-switched ruby laser, which delivered 1 J in 25 ns, was split and one component used to produce a plasma of the ion species to be studied while the other generated a back-lighting continuum. The feasibility of the method depends on the availability of intense, line free continua emitted from plasmas produced on appropriate high-Z targets.[3] The system was employed to detect and study the autoionizing resonances of Li^+, but had a limited range of time resolution which was based purely on the use of optical-path differences.

In the modified experiment described here, two Q-switched lasers were used. The ruby laser generated the absorbing plasma while a neodymium-glass oscillator-amplifier system, which gave a 3-J, 20-nsec output pulse at 1060 nm, was used to produce the continuum from a tungsten target. The pumping pulses in the two laser systems were synchronized electronically to peak simultaneously and a fraction of the ruby output was used to trigger the drive unit of the neodymium laser's Pockels cell. A schematic diagram of the apparatus is given in Fig. 1. With this arrangement a continuously variable delay from 250 nsec to 10 msec could be introduced between the two output pulses. An electronic circuit linking the two Q-switching units and enabling the lasers to be fired in either order with delay times from 0 to 100 msec has also been developed.

Although designed primarily for ionic absorption studies, the system, because of its flexibility, is also applicable to the investigation of neutral atoms. Thori-

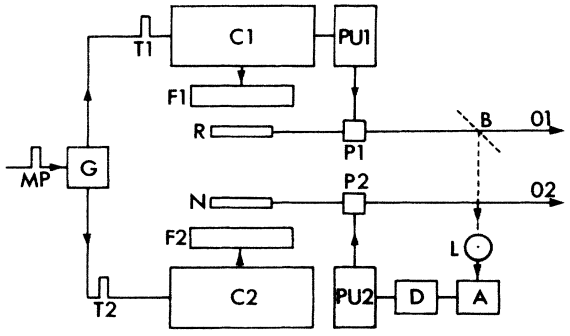

FIG. 1. Schematic diagram of dual-laser absorption physics. MP, master pulse; G, delayed pulse generator; T1, trigger pulse to ruby-laser control circuit; T2, trigger pulse to neodymium oscillator-amplifier system; C1,C2, charging and control circuits; F1,F2, flash lamps; R, ruby rod; N, neodymium oscillator-amplifier rods; P1,P2, Pockels cells; PU1,PU2, Pockels-cell control units; B, beam splitter; L, light sensitive diode; A, amplifier; D, delay circuit; O1,O2, optical output pulses.

um was chosen as an interesting candidate for study because not only is it challenging by being very refractory (mp, 1750 °C; bp, ~4790 °C) and extremely corrosive, but also its absorption spectrum in the extreme is of current theoretical importance as a test of the application of random-phase approximation (RPA) methods to collective resonances in the actinides.[4,5] To date only the spectra of the metal[6] and the compound ThF_4[7] have been reported, and comparison with theory has been made on the presumption that the atomic profiles will not be seriously modified by the metallic environment.

To generate neutral species in our system it is necessary to (1) irradiate the metal target with a largely defocused laser beam so that the irradiance is small and a plasma of low ion stage is generated, and (2) use a long time delay between the two laser pulses. In the absorbing plasma system the irradiated area was ~6 mm in diameter corresponding to a peak power density of 2×10^9 W/cm^2. According to the collisional-radiative theory of Colombant and Tonan,[8] the resulting electron temperature in the plasma would be 3 to 5 eV and the dominant ion species Th^{++}. A plasma in such initial ion stages should rapidly revert to a neutral system. The time delays used in our observations varied from 500 to 800 nsec which should be adequate for total or near-total recombination. As a check on this assumption, spectra of samarium and lanthamum were taken under identical conditions and were found to be very similar to those obtained by Radtke,[9] except for a weak absorption feature to the long-wavelength side of the main maximum in lanthanum and the suggestion of a shoulder at higher energies which may have been due to residual singly ionized lanthanum ions.

The spectra were photographed on a 2-m grazing-incidence vacuum spectrograph with a 1200-lines/mm Bausch and Lomb grating. With a slit width of 100 μm, 300 laser shots were required to obtain a good spectrum on Kodak SWR plates. Lines from a beryllium oxide target were used as references to calibrate the spectra.

In the past decade or so much work, both experimental[10-14] and theoretical,[15-20] has been reported on the $4d$-f spectra of the lanthanides. Experimentally the spectra consist of intense broad absorption resonances, typically some tens of electron volts wide and exhibiting several peaks. Theoretically they are best explained in terms of many-body calculations,[15,18,20] and the application of the random-phase approximation with exchange by Wendin[19] to the case of lanthamum gave excellent agreement with the experimental cross sections of Radtke.[9] More recently Wendin[4] has applied a modified form of RPA—the local-density random-phase approximation—to photoionization in thorium and uranium. Near the $5d^9\,^2D$ limits the theory predicts a giant dipole resonance with a two-peak structure for both thorium and uranium [Fig. 2(a)]. In the calculation the contribution of the outer unfilled shells to the ground-state potential is introduced as a spherical average. From one simple viewpoint, therefore, the spectrum may be considered as arising from the $5d^{10}5d^95f$ core excitation which, because of the magnitude of the $5d$-$5f$ overlap, has a large transition moment and a large exchange interaction.[21] The weaker and narrower peak of longer wavelength may be associated with the intersystem $^1S_0$-$^3P_1,^3D_1$ transitions while most of the oscillator strength goes into the broad $^1S_0$-$^1P_1$ transition which, because of the large exchange integral, is moved by about 13 eV to higher energies.

The spectrum of thorium vapor obtained in the present experiment is shown in Fig. 2 where it is compared with (a) Wendin's theoretical profile,[4] (b) the metal spectrum,[6] and (c) the spectrum of ThF_4.[7] The experimental profile is based on microdensitometer traces corrected for background and normalized so that the height of the main resonance peak is the same as in Wendin's theoretical results. It can be seen from the figure that, although we do not have absolute experimental cross sections for comparison, there is good general agreement between the theoretical predictions and the absorption curve for the vapor. Thus the observed position of the main peak coincides within experimental error with the theoretical prediction. The longer-wavelength peak is shifted slightly (by about 2 eV) to higher energies relative to the theoretical peak, is broader, and has a lower maximum. That further broadening would occur is in this peak as a result of $5d$ Auger processes and the effects of $5f$ hybridization was recognized by Wendin.[4] The dip in the theoretical curve at 100 eV arises from interaction with $5d^97p$ excitation; it might be expected to appear in vapor absorption but was not detected in our experiments.

In Fig. 2(b) we compare our atomic results with those for the metal.[6] The remarkable similarity of the spectra is evidence that the potential of the $5f$ electron is largely unaffected by its environment in the solid. In Fig. 2(c) the present results are compared with those reported for ThF_4 vapor[7] and again it is seen that the environment, in this case the fluorine cage of the molecule, does not seriously modify the core wave functions.

In the similarity of its atomic and metal spectra, thorium bears a strong resemblance to lanthanum. In proceeding up the periodic table lanthanum is the last element with a empty $4f$ shell. According to Radtke[10] vapor-metal shifts in the lanthanides occur in those cases when in the process of metal formation a $4f$ electron is displaced out of the core so that the metal spectrum of element Z resembles that of the vapor spectrum of element $Z-1$; clearly this cannot happen in

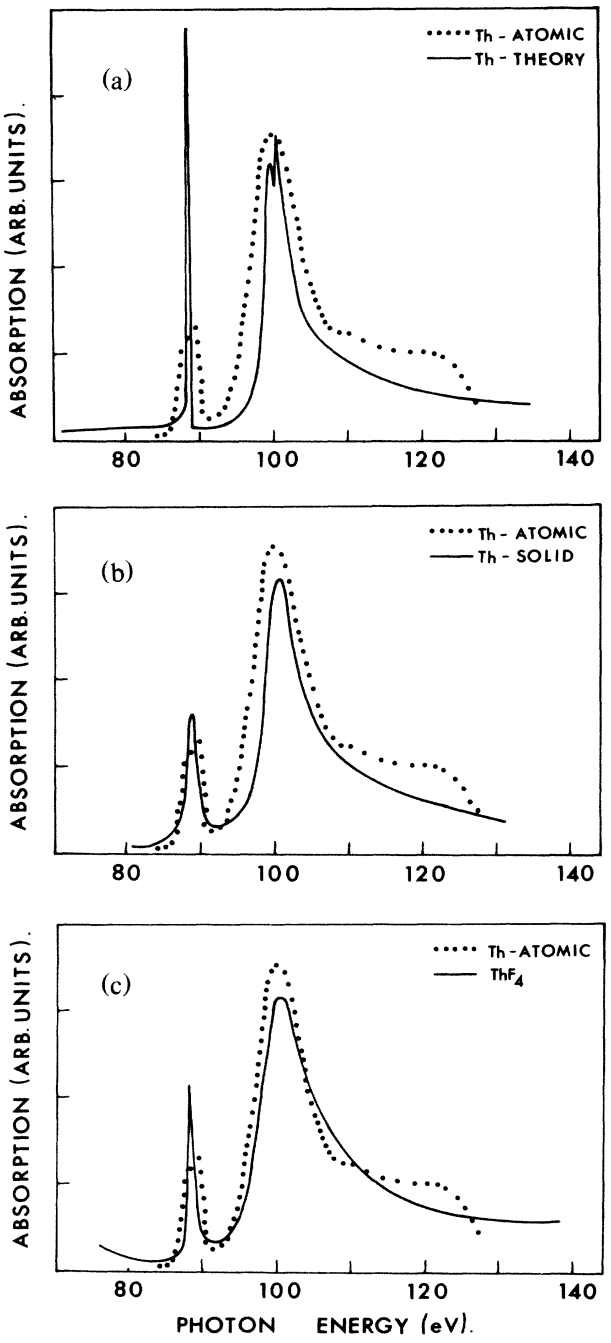

FIG. 2. Photoionization of Th($7s^26d^2$) near the $5d$ limit. (a) Comparison of present results with local-density random-phase approximation theory of Wendin (Ref. 4). The experimental results are scaled so that the height of the main peak coincides with the height in Wendin's curve. (b) Comparison of present results with the photoionization cross section of thorium metal (Ref. 6) as given by Wendin (Ref. 4). (c) Comparison of present results with cross section (Ref. 7) for ThF_4. The shoulder structure on the high-energy side of our traces may originate from residual ionic absorption or may be simply an artifact of our experimental procedure.

lanthanum. In a similar way thorium is the last element with an empty $5f$ shell so that the possibility of $5f$ excitation on metal formation, with a consequent spectral shift, again does not arise. Thus the present results provide the first example of what might be expected for the systematics of vapor-metal spectral shifts in the actinides.

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