

The new measurement reported here agrees with the theoretical value for this interval and is clearly competitive with the other measurements.

It is clear from Table I that the primary limitation in the measurement of the  $2^2P_{3/2}$ - $2^2S_{1/2}$  fine-structure interval is the systematic error and not the statistical error. Of the systematic errors the most troublesome is the variation in the relative phase of the microwave fields in the two waveguides when the apparatus is taken apart and reassembled. It is our conclusion from a separate measurement of the Lamb shift that these systematic errors can be reduced through improvements in the apparatus, the procedure used to take data, and further theoretical studies of the resonance line shape. This should conservatively yield an order-of-magnitude increase in the precision of the determination of this fine-structure interval to the level of 0.4 ppm. This would provide a value of the fine-structure constant to 0.2 ppm or, alternatively, with use of  $\alpha$  determined from other sources, a measurement of the Lamb shift to 4 ppm.

We would like to acknowledge the support of the National Science Foundation through Grant No. 78-09657.

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<sup>2</sup>For a report on the use of the separated-oscillatory-field technique to measure the Lamb shift in  $H(n=3)$ , see C. W. Fabjan and F. M. Pipkin, *Phys. Rev. A* **6**, 556 (1972).

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## Observation of the Auger Resonant Raman Effect

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(Received 13 August 1980)

Monochromatized synchrotron radiation near the photoionization threshold was used to produce the  $2p_{3/2}^{-1}$  vacancy state in atomic Xe. Deexcitation of the state through  $L_3$ - $M_4M_5(^1G_4)$  Auger-electron emission was measured. The  $5d$  spectator-electron Auger satellite was observed. The satellite energy exhibits linear dispersion as a function of the photon energy of the exciting radiation. The observed width of the  $^1G$  diagram line decreases by  $\sim 40\%$  at threshold. This radiationless process can thus be construed as the Auger analog of the x-ray resonant Raman effect.

PACS numbers: 32.80.Hd, 32.70.Fw

The discovery of x-ray resonant Raman scattering<sup>1-3</sup> suggests the existence of an analogous radiationless process. In the x-ray resonant

Raman effect, absorption of a photon  $\hbar\omega_1$  promotes an atomic inner-shell electron to an excited bound state; the inner-shell vacancy is

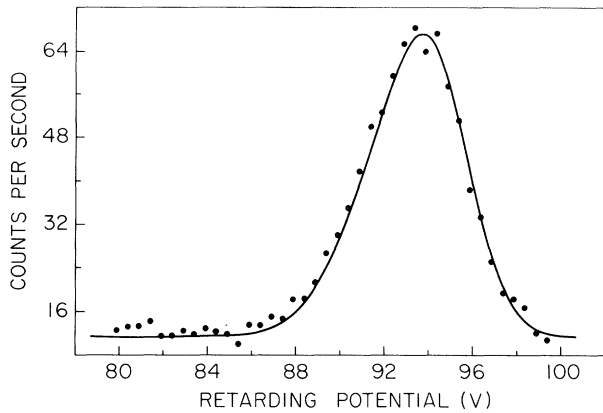


FIG. 1. Xenon  $L_3$  photoelectron spectrum, excited with  $\sim 5$ -keV x rays.

simultaneously filled by another atomic electron under emission of a characteristic x-ray photon  $\hbar\omega_2 (< \hbar\omega_1)$ . Absorption and emission occur in a single process, described by taking the  $\vec{p} \cdot \vec{A}$  term in the interaction Hamiltonian to second order in perturbation theory.<sup>3</sup> Consequently, the atom "remembers" how it was excited: The resonantly emitted x-ray line reflects the width of the exciting radiation, *not only* the natural lifetime width  $\Gamma$  of the inner-shell hole state (just as in resonance fluorescence<sup>4</sup>). If excitation is by a sharp line, then  $\hbar\omega_2$  varies in energy with  $\hbar\omega_1$  over a range  $\sim \Gamma$ . This linear dispersion and the narrowing of the scattered radiation are the

identifying characteristics of the resonant Raman process.

We have employed synchrotron radiation to ionize the  $L_3$  shell of Xe near threshold and have examined the subsequent  $L_3$ - $M_{4,5}$  $M_{4,5}$  Auger transitions, in search of a resonant Raman effect in which the atom's deexcitation occurs through a radiationless, rather than radiative, transition. The experiment was performed on a focused x-ray beam at the Stanford Synchrotron Radiation Laboratory. A doubly curved gold-coated mirror situated 11.5 m from the source condensed 2.5 mrad of synchrotron radiation onto the target gas jet. The radiation was monochromatized by two germanium crystals arranged in the parallel, (111) symmetric configuration. The experiment was performed at 3.1 GeV electron-beam energy and 60 mA mean current, yielding a photon flux of  $\sim 5 \times 10^{10}$  photons/sec through a  $2 \times 4$ -mm<sup>2</sup> aperture upstream of the target. The full width at half maximum (FWHM) of the incident x-ray spectrum at the Xe  $L_3$  edge (4786 eV) was  $\sim 2.5$  eV, mainly due to the vertical angular divergence of the source.

The target consisted of a gas jet formed by a glass capillary (0.2 mm i.d.); the jet intersected the x-ray beam at right angles in the horizontal plane. The pressure in the interaction region was calculated to be  $\sim 0.1$  Torr, yielding a background pressure in the vessel of  $5 \times 10^{-4}$  Torr.

The photoelectrons were analyzed by a com-

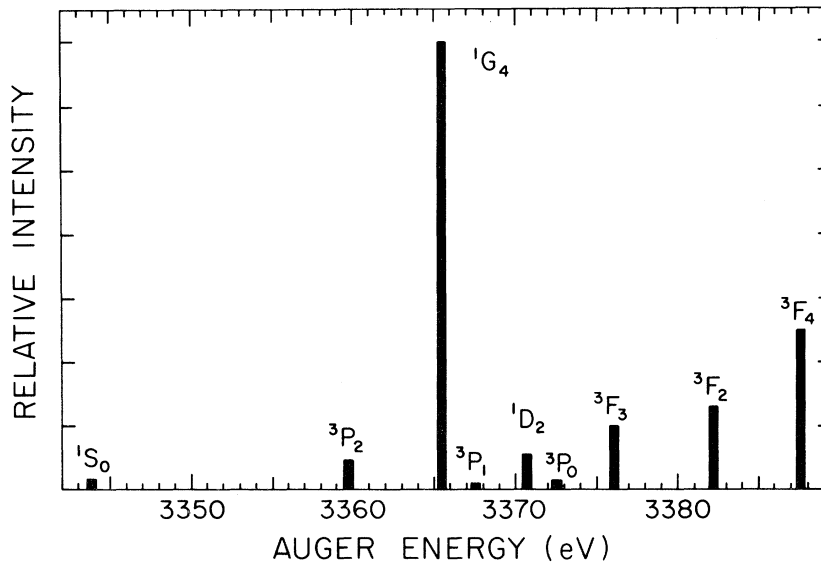


FIG. 2. The Xe  $L_3$ - $M_{4,5}$  $M_{4,5}$  Auger spectrum, calculated *ab initio* with Dirac-Hartree-Slater wave functions, in intermediate coupling.

mercial double-pass cylindrical-mirror analyzer with its symmetry axis in the vertical plane. The spectrometer energy resolution was  $\sim 2.5$  eV FWHM. The stability of the overall system (x-ray monochromator and electron spectrometer) was checked frequently by measuring the position and width of the Xe  $L_3$  photoelectron peak excited with  $\sim 5$ -keV x rays (Fig. 1). The photopeak is 5.4 eV wide (FWHM), corresponding to a Voigt-function convolution<sup>5</sup> of the  $\sim 3.0$ -eV-wide  $2p_{3/2}^{-1}$  hole-state Lorentzian with the 2.5-eV spectrometer transmission function and the 2.5-eV-wide incident spectrum.

The immediate vicinity of the  $L_3$ - $M_4M_5(^1G_4)$  Auger-electron line was scanned. This line arises from the deexcitation of  $\sim 18\%$  of all  $2p_{3/2}^{-1}$  vacancies (Fig. 2). When the Xe atoms are ionized with photons well ( $\sim 200$  eV) above the  $L_3$  binding energy, the measured  $^1G$ -line spectrum has a Lorentzian shape of  $\sim 6.1$  eV FWHM. As the incident photon energy is lowered to and below threshold, the  $^1G$  diagram line is observed to shift to higher energy. Above the diagram line, a spectator satellite appears (Fig.

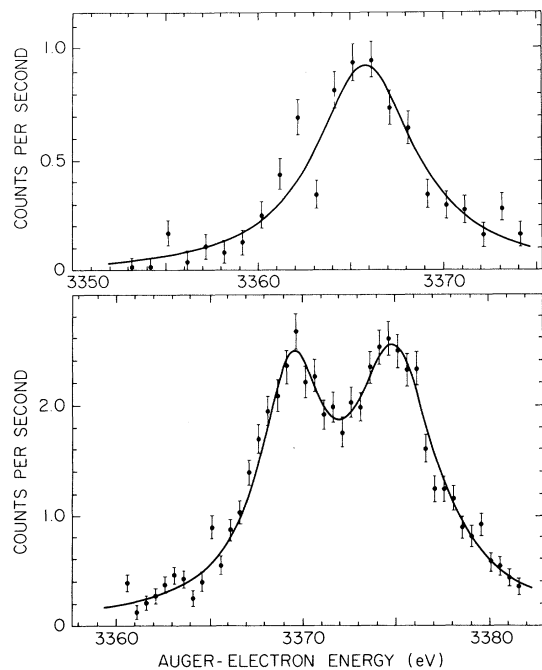


FIG. 3. The Xe  $L_3$ - $M_4M_5(^1G_4)$  Auger line, from atoms excited  $\sim 200$  eV above threshold (top), and from atoms excited 2.0 eV below threshold (bottom). In the latter spectrum, the  $^1G$  diagram line is accompanied on the high-energy side by the  $5d$  spectator-electron satellite. The Lorentzian fits are characterized by  $\chi^2 \leq 1$ .

3) that persists even after the intensity of the diagram line has vanished some 5 eV below threshold.

In Fig. 4(a), the measured Xe  $L_3$  absorption edge is analyzed in terms of the  $2p_{3/2}$ -electron transition probability to the continuum and to unoccupied bound  $6s$ ,  $5d$ ,  $6d$ , and  $7d$  states.<sup>6</sup>

Auger energies are plotted in Fig. 4(b) against

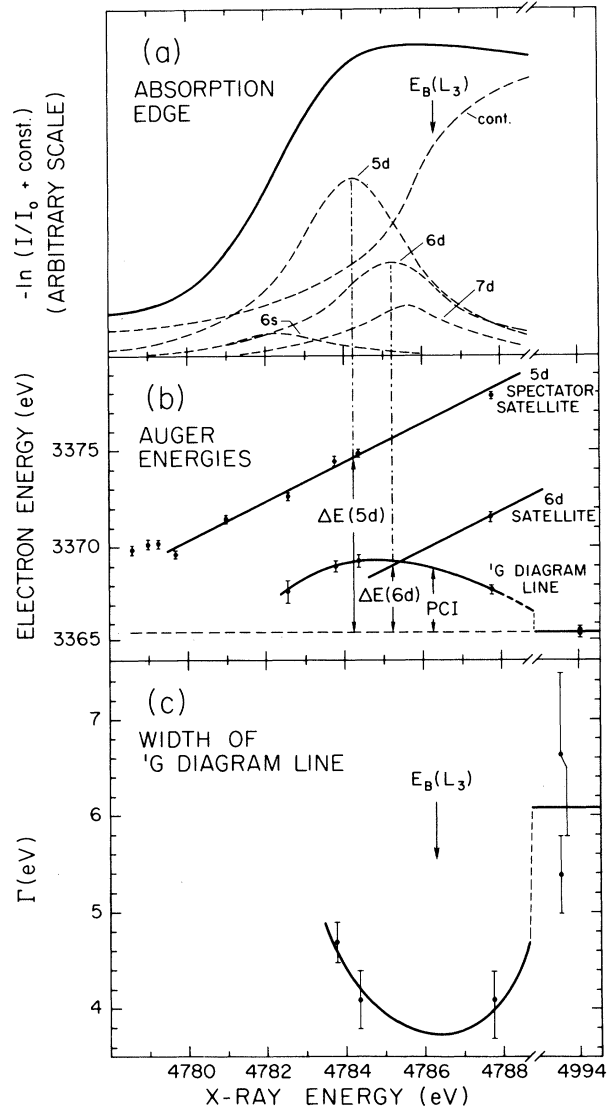


FIG. 4. (a) Measured Xe  $L_3$  absorption edge, decomposed according to Ref. 6. (b) Energies of the  $L_3$ - $M_4M_5(^1G_4)$  Auger line and its satellites. Near threshold, the  $^1G$  diagram line is shifted by post-collision interaction (PCI). The satellites are shifted due to screening by  $5d$  and  $6d$  spectator electrons, respectively; their energies exhibit linear (Raman) dispersion. (c) Width of the measured  $^1G$  diagram-line spectrum, as a function of excitation energy.

incident photon energy. The  $\sim +3$  eV shift of the  ${}^1G$  diagram line, near threshold, is presumably due to post-collision interaction (PCI),<sup>7,8</sup> i.e., the influence of the Coulomb field of the slowly receding photoelectron. (A similar, much smaller shift has been observed in the Xe  $N_{4,5}$ -OO Auger spectrum.<sup>8</sup>) The magnitude of the present PCI shift is in accord with the predictions of the Niehaus theory.<sup>7,9</sup>

The  $5d$  spectator-electron satellite exhibits the characteristic Raman linear dispersion,<sup>2</sup> while the expected dispersion of the diagram line is masked by the PCI. The  $5d$  satellite is shifted by 9 eV when the atom is excited at the centroid energy of the  $2p_{3/2} \rightarrow 5d$  transition. The theoretical prediction for this shift is 7.2 eV, from a calculation with our relativistic relaxed-orbital Auger-energy code.<sup>10</sup> In addition to the  $5d$  satellite, a  $6d$  spectator-electron satellite is observed in an Auger spectrum excited at  $h\nu = 4787.8$  eV. It is reasonable to assume the same dispersion for the  $6d$  satellite as for the  $5d$  satellite, because the width of both transition-probability functions is governed by the  $L_3$ -hole width. We then deduce a 3.5-eV shift of the  $6d$  satellite with respect to the  ${}^1G$  diagram line (without PCI) at the centroid of the  $2p_{3/2} \rightarrow 6d$  transition probability [Fig. 4(b)], in agreement with a theoretical shift of 3.3 eV calculated with the relativistic Auger-energy code.<sup>10</sup>

The measured width of the  ${}^1G$  diagram line is plotted in Fig. 4(c) as a function of exciting photon energy. The data indicate a narrowing by  $\sim 40\%$  of the Auger diagram line when the  $2p_{3/2}^{-1}$  hole state is excited at threshold. This effect is analogous to the narrowing below lifetime width observed in x-ray resonant Raman scattering.<sup>2</sup>

The width of the  $5d$  satellite line, by contrast, remains constant at  $5 \pm 1$  eV over the entire excitation-energy range covered by these experiments. Unresolved multiplet splitting is expected to account for the broadening of this line.

We thank Marianne Breinig, M. O. Krause, C. W. Nestor, Fernando Parente, and F. Wuilleumier for helpful discussions. This work was supported by the U. S. Air Force Office of Scientific Research and the U. S. Army Research Office. Materials incorporated in this work were developed at the Stanford Synchrotron Radiation Laboratory, which is supported by the National Science Foundation in cooperation with Stanford Linear Accelerator Center and the U. S. Department of Energy.

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