Open access • Journal Article • DOI:10.1103/PHYSREVLETT.45.1937

Observation of the Auger Resonant Raman Effect — Source link

George Brown, Mau Hsiung Chen, Bernd Crasemann, Gene E. Ice

Institutions: Stanford University, University of Oregon, Oak Ridge National Laboratory

Published on: 15 Dec 1980 - Physical Review Letters (American Physical Society)

Topics: Auger effect, Auger electron spectroscopy, Binding energy, Raman scattering and Photoionization

CONF-8011129--1

DE82 004088

ted t
Letter

Observation of the Auger Resonant Raman Effect*

George S. Brown
Stanford Synchrotron Radiation Laboratory
Stanford, California 94305

Mau Hsiung Chen and Bernd Crasemann
Department of Physics, University of Oregon
Eugene, Oregon 97403

Gene E. Ice
Metals and Ceramics Division, Oak Ridge National Laboratory+
Oak Ridge, Tennessee 37830

Monochromatized synchrotron radiation near the photoionization threshold was used to produce the $[2p_{3/2}]$ 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. The observed width of the ^1G diagram line decreases by ~40% when the exciting photon energy reaches the vicinity of the Xe L_3 binding energy. This radiationless process can thus be construed as the Auger analog of the x-ray resonant Raman effect. The ^1G diagram line is shifted by ~+3 eV due to post-collision interaction; this shift varies with excitation energy.

PACS numbers: 32.80.Hd, 32.70.Fw

*This work was supported by the Air Force Office of Scientific Research and the U.S. Army Research Office.

+Operated by Union Carbide Corporation under contract W-7405-eng-26 with the U.S. Department of Energy.

The discovery of x-ray resonant Raman scattering[1-3] suggests the existence of an analogous radiationless process. We have employed synchrotron radiation to ionize the L_3 shell of Xe near threshold, and have examined the subsequent deexcitation through L_3-$M_{4,5}M_{4,5}$ Auger transitions.

The experiment was performed on a focussed 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 ~5×10^{10} photons/sec through a 2x4-mm 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 ~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 (0.1-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 ~0.1 Torr, yielding a background pressure in the vessel of 5×10^{-4} Torr.

The photoelectrons were analyzed by a commercial double-pass cylindrical mirror analyzer with its symmetry axis in the vertical plane. The spectrometer energy resolution was ~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 (Fig. 1), which was 5.4 eV wide (FWHM), corresponding to a Voigt-function convolution[4] of the ~3.0-eV wide $2p_{3/2}$ 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 ~18% of all $2p_{3/2}$ vacancies (Fig. 2). When the Xe atoms are ionized with photons well (~100 eV) above the L_3 binding energy, the measured ^1G-line spectrum has a Lorentzian shape of ~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. 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.[5]

Auger energies are plotted in Fig. 4(b) against incident photon energy. The ~+3-eV shift of the ^1G diagram line, near threshold, is presumed to be due to post-collision interaction (PCI).[6] No theory of PCI below threshold exists to date to our knowledge, whence the energy dependence of the effect must await explanation. The 5d spectator-electron satellite exhibits the characteristic Raman linear dispersion,[2] 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 $2p_{3/2}$→5d transition. The theoretical prediction for this shift is 7.2 eV, from a calculation with our relativistic relaxed-orbital Auger-energy code.[7] In addition to the 5d satellite, a 6d spectator-electron satellite is observed in an Auger spectrum excited at hν=4787.3 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.[7]

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 ~40% of the Auger diagram line when the $[2p_{3/2}]$ hole state is excited at threshold. This effect is analogous to the narrowing below lifetime width observed in x-ray resonant Raman scattering.[2] The width of the 5d satellite line, by contrast, remains constant at (5±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, and Fernando Parente for helpful discussions. This work was supported by the 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 SLAC and the Department of Energy.

[1]C. J. Sparks, Phys. Rev. Lett. 33, 262 (1974).

[2]P. Eisenberger, P. M. Platzman, and H. Winick, Phys. Rev. Lett. 36, 623 (1976), and references therein.

[3]P. Suortti, Phys. Stat. Sol. (b) 91, 657 (1979).

[4]O. Keski-Rahkonen and M. O. Krause, Phys. Rev. A 15, 959 (1977).

[5]M. Breinig, M. H. Chen, G. E. Ice, F. Parente, B. Crasemann, and G. S. Brown, Phys. Rev. A 22, 520 (1980).

[6]A. Niehaus, J. Phys. B 10, 1845 (1977).

[7]M. H. Chen, B. Crasemann, K.-N. Huang, M. Aoyagi, and H. Mark, At. Data Nucl. Data Tables 19, 97 (1977).

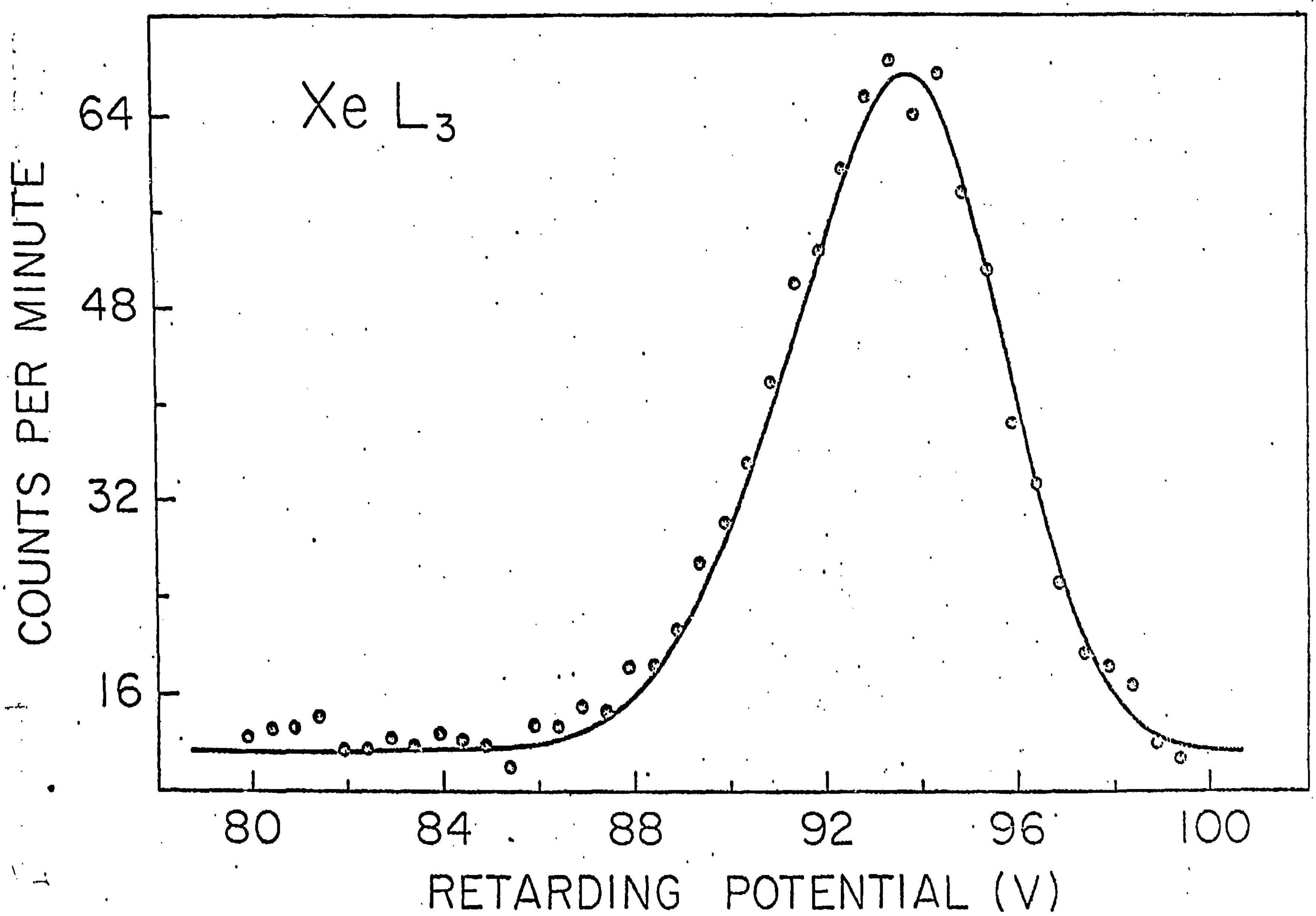
Figure Captions

FIG. 1. Xenon L_3 photoelectron spectrum.

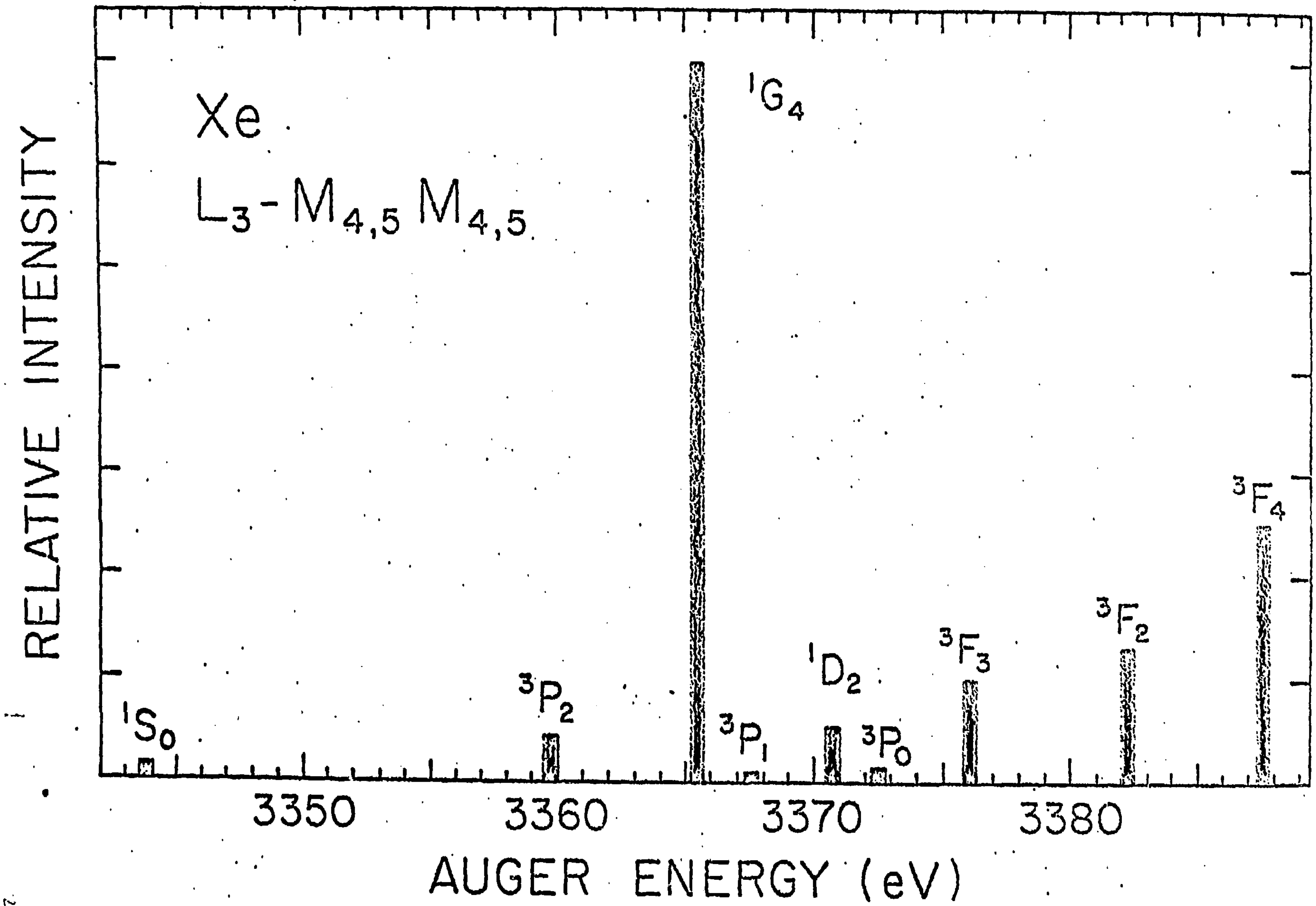
FIG. 2. The Xe L_3-$M_{4,5}M_{4,5}$ Auger spectrum, calculated *ab initio* with Dirac-Hartree-Slater wave functions, in intermediate coupling.

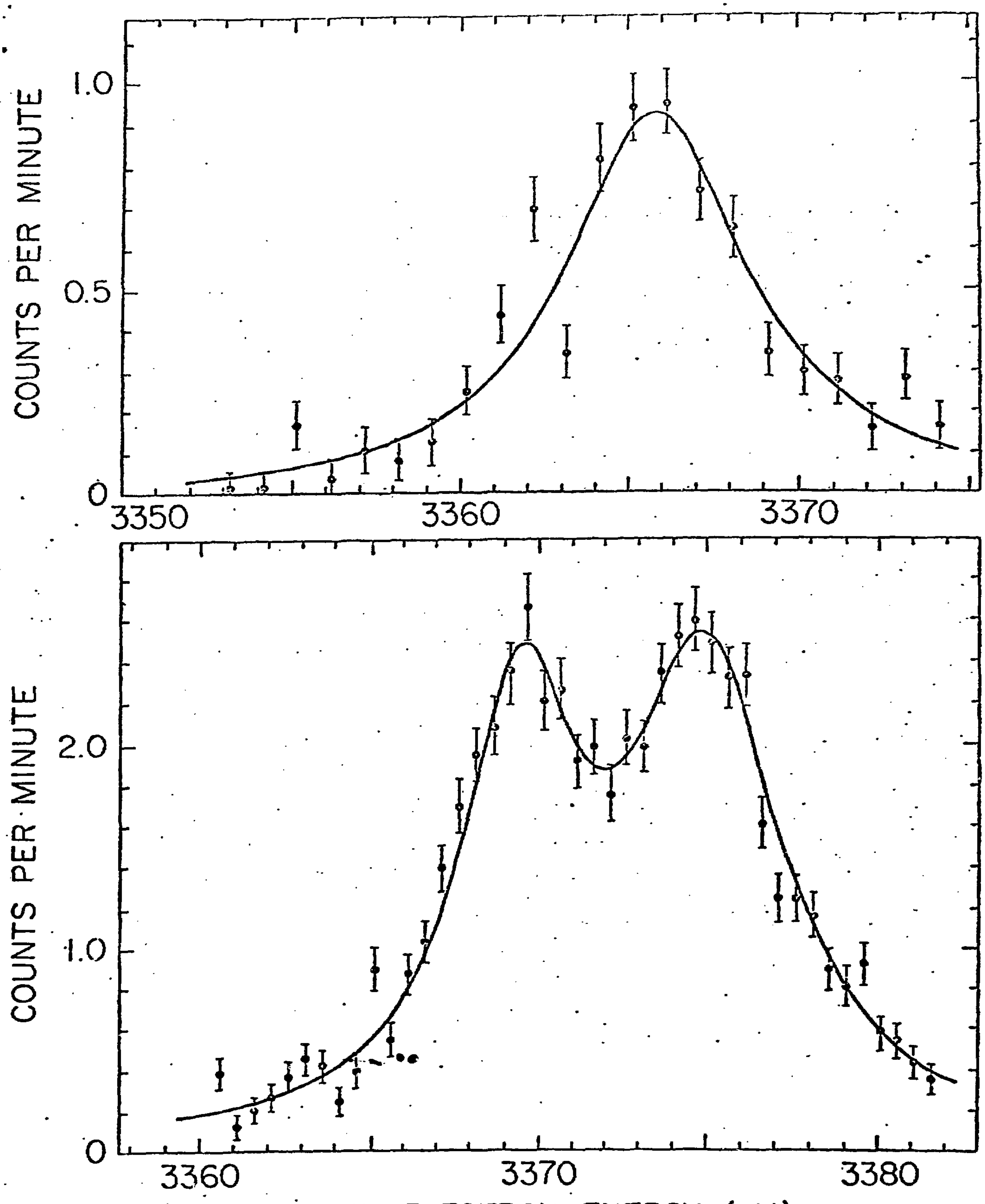
FIG. 3. The Xe L_3-$M_4M_5(^1G_4)$ Auger line, from atoms excited ~100 eV above threshold (*top*), and from atoms excited 2.0 eV below threshold (*bottom*). In the latter spectrum, the ^1G diagram line is accompanied on the high-energy side by the 6d spectator-electron satellite. The Lorentzian fits are characterized by $\chi^2 = \leq 1$.

FIG. 4. (a) Measured Xe L_3 absorption edge, decomposed according to Ref. 4. (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.



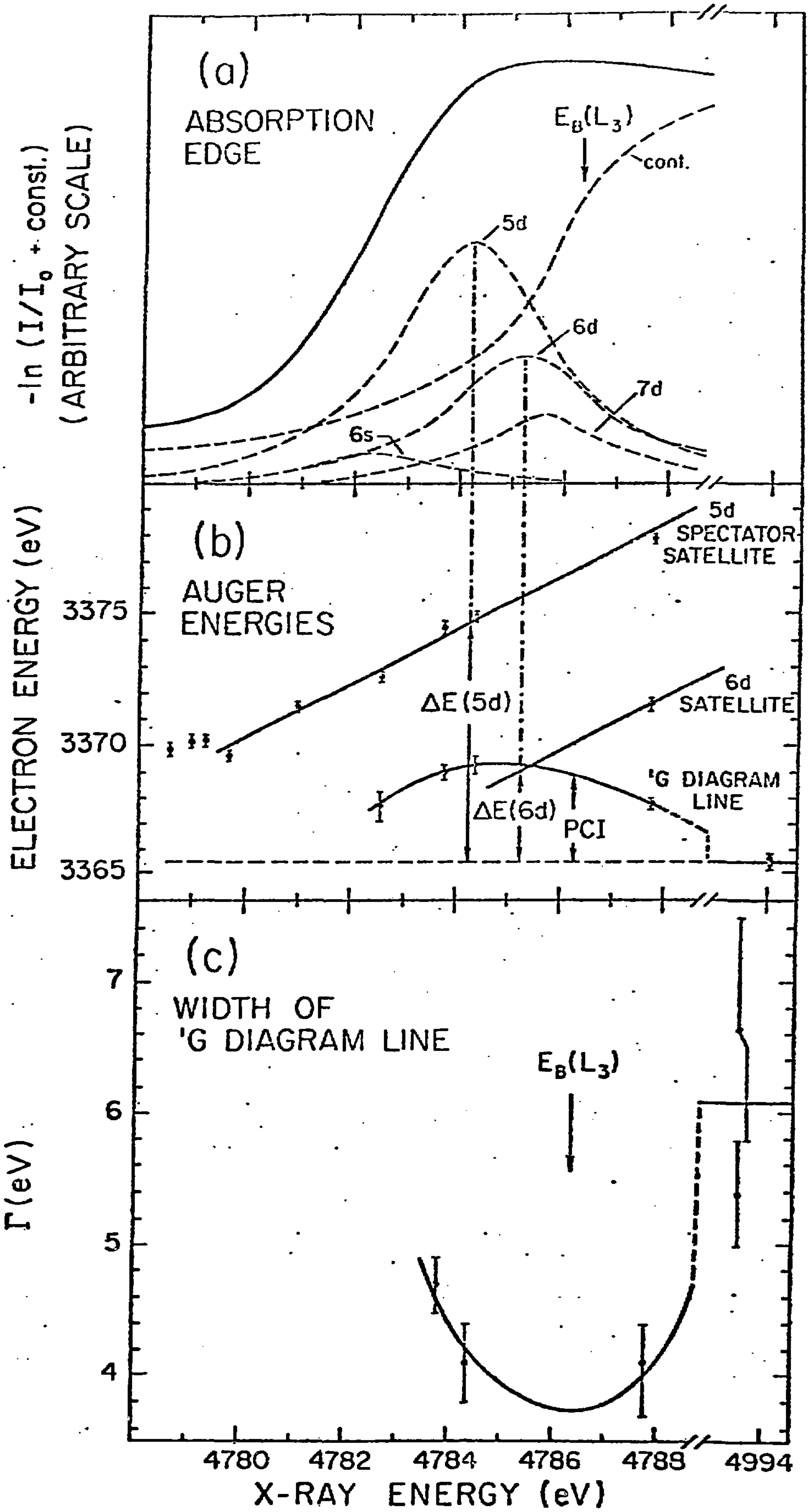
Xe L_3
64
48
32
16
COUNTS PER MINUTE
80
84
88
92
96
100
RETARDING POTENTIAL (V)


Xe
L3-M4,5 M4,5
RELATIVE INTENSITY
AUGER ENERGY (eV)
3350
3360
3370
3380
1S0
3P2
1G4
3P1
1D2
3P0
3F3
3F2
3F4



COUNTS PER MINUTE
1.0
0.5
0
3350
3360
3370
COUNTS PER MINUTE
2.0
1.0
0
3360
3370
3380
AUGER-ELECTRON ENERGY (eV)

Fig. 3



(a)
ABSORPTION EDGE
$E_B(L_3)$
cont.
5d
6d
7d
6s
-ln (I/I_o + const.) (ARBITRARY SCALE)
(b)
AUGER ENERGIES
5d SPECTATOR SATELLITE
6d SATELLITE
'G DIAGRAM LINE
ΔE(5d)
ΔE(6d)
PCI
ELECTRON ENERGY (eV)
3375
3370
3365
(c)
WIDTH OF 'G DIAGRAM LINE
$E_B(L_3)$
Γ(eV)
7
6
5
4
4780
4782
4784
4786
4788
4994
X-RAY ENERGY (eV)

Fig. 4