of these resonances is given by the $^{12}C+^{12}C$ and α_0 widths, Γ_C and Γ_{α_0}. From $\sigma_{tot}=8\pi\lambda^2(2L+1)\Gamma_C \times\Gamma_{\alpha_0}/\Gamma^2=8.9\pm2.3$ mb for all the 11.2-MeV 8^+ resonance, one obtains $\Gamma_C\Gamma_{\alpha_0}/\Gamma^2=0.015\pm0.004$, which is comparable with the value of 0.0068 obtained in Ref. 2 for the 19.0-MeV 10^+ resonance. Furthermore, taking $\Gamma\sim\Gamma_{\alpha_0}+\Gamma_C$, one obtains that $\Gamma_{\alpha_0}\sim\Gamma_C$ within the experimental error. From the relative penetrabilities of ^{12}C and α particles, for the reduced widths we conclude $\gamma_C^2>\gamma_{\alpha_0}^2$, thus strengthening the "molecular" picture of the resonances.

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Three-Photon Excitation of Xenon and Carbon Monoxide

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With intense light of a pulsed tunable dye laser, the $^3P_1$ resonance state of xenon and the P, Q, and R branches of the (2-0) band of CO in the fourth positive system ($A^1\Pi\leftarrow X^1\Sigma^+$) are excited by simultaneous absorption of three visible laser photons. The excitation is monitored by detecting the fluorescent decay in the vacuum ultraviolet.

We report the excitation of Xe and CO by simultaneous absorption of three visible photons. In this first observation of the excitation of an atomic and a molecular gas by three-photon absorption[1] the $^3P_1$ resonance state of xenon and the P, Q, and R branches of the (2-0) band of CO in the fourth positive system ($A^1\Pi\leftarrow X^1\Sigma^+$) are excited with intense tunable-dye-laser radiation of about 4400 Å. The three-photon excitation is monitored by detecting the fluorescent decay of the excited states in the vacuum ultraviolet.

There is considerable interest in three-photon excitation of atomic and molecular states. For instance, as demonstrated in the present experiments, this method permits high-resolution laser spectroscopy in the spectral region of the vacuum ultraviolet in a simple experimental arrangement. In this spectral region nonlinear laser spectroscopy has been performed so far by two-photon excitation of atoms[2] and molecules[3] with intense ultraviolet light of frequency-doubled dye lasers. Since two-photon excitation is restricted to the excitation of levels of the same parity as the ground state and three-photon absorption excites states of opposite parity, the two methods complement each other for nonlinear spectroscopy. As discussed below, both methods can provide a Doppler-free recording of the observed transitions in an almost identical, simple scheme of excitation.

The intense tunable-laser radiation, required for the present experiments, is generated with a pulsed-dye-laser oscillator amplifier system.[4] For the recording of the three-photon exitation of xenon (Fig. 1) the blue laser light, exceeding 80-kW pulse peak power, was focused by a lens of 500-mm focal length into a small gas cell (a 50-cm-long section of 20-mm-diam glass tubing with parallel quartz windows) to a beam waist of about 50 μm. The fluorescence at $\lambda=1470$ Å is observed through a MgF_2 side window and monitored by a solar-blind photomultiplier (EMR 541 G). The signal is amplified and accumulated by a gated integrator before being recorded by an x-y recorder. Because of the high transition probability[5] ($A=2.2\times10^8$ sec^{-1}) of the observed resonance transition, the fluorescence is resonantly trapped and reduced due to the quenching

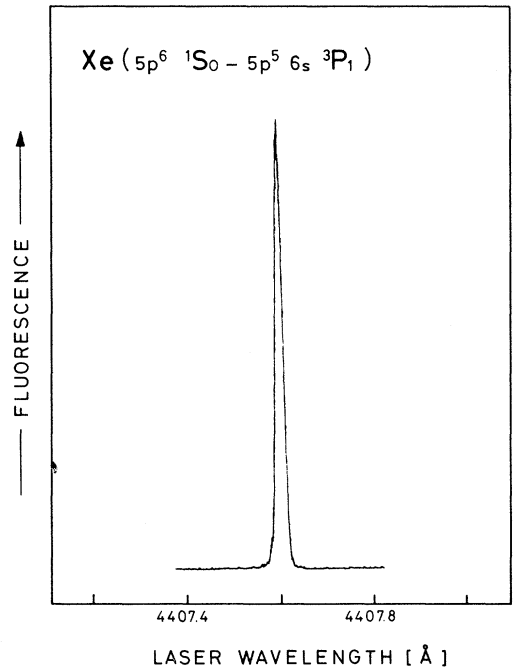

FIG. 1. Three-photon–excited resonance fluorescence in Xe at 1470 Å, recorded at a rate of 0.02 Å per minute and an effective time constant of $\tau = 10$ sec. Xe pressure is 8 mTorr.

of excited atoms by gas impurities. To minimize this effect the distance between the light focus and the inner convex surface of the MgF_2 window was adjusted to be less than 50 μm. The maximum fluorescence was observed at pressures between 2 and 8 mTorr.

The transition probability W_f for a three-photon absorption from the ground state $|g\rangle$ to the excited state $|f\rangle$ can be obtained from lowest-order perturbation theory[6-8] as, $W_f = 2\pi[I(\omega)/I](2\pi\alpha I)^3 |K_f|^2$ with

$$K_f = \sum_{m,n} \frac{\langle f|\vec{\epsilon}\cdot\vec{r}|m\rangle\langle m|\vec{\epsilon}\cdot\vec{r}|n\rangle\langle n|\vec{\epsilon}\cdot\vec{r}|g\rangle}{(\omega_m - 2\omega)(\omega_n - \omega)}.$$

In these equations, I is the laser intensity and $I(\omega)$ is the intensity per unit bandwidth. The frequencies ω, ω_n, and ω_m refer to the frequency of the laser and to the eigenfrequencies of the atomic states $|n\rangle$ and $|m\rangle$. The summations run over the complete set of states (including the continuum) of the atomic Hamiltonian.

For Xe an exact evaluation of K_f is not yet practicable because the wave functions and hence the required matrix elements are not known. For the present experiment, however, an order-of-magnitude estimate should be obtainable by using the average-frequency method of Bebb and Gold.[7] In terms of the average frequency[9] $\bar{\omega}$, K_f reduces to

$$K_f = \frac{\langle f|(\vec{\epsilon}\cdot\vec{r})^3|g\rangle}{(\bar{\omega} - 2\omega)(\bar{\omega} - \omega)}.$$

For Xe the matrix element $\langle {}^3P_1|(\vec{\epsilon}\cdot\vec{r})^3|{}^1S_0\rangle$ is estimated to be $0.15a_0^3$ (a_0, Bohr radius) using a Coulomb approximation. This is consistent with the oscillator strength $f = 0.21$ of this transition.[5]

From the equations above we estimate that under the experimental conditions (power density $d = 4$ GW/cm^2, laser bandwidth $\Delta\omega_L = 1.5$ GHz, interaction volume $V = 1.2\times10^{-7}$ cm^3, Xe pressure $p = 8$ mTorr) more than 300 atoms should be excited during the laser pulse.

The fluorescent intensity observed at the center of the resonance (Fig. 1) corresponds, on the average, to about 2.5 detected photons per laser pulse. Taking the detection solid angle (2%), the quantum efficiency of the photomultiplier (21%) and the transmission of the MgF_2 window (70%) into account, we conclude that about 850 xenon atoms are excited by each laser pulse. This number can be considered to be in approximate agreement with the theoretical order-of-magnitude estimate.

The signal-to-noise ratio and low background signal, which is obtained for the recorded resonance (Fig. 1) despite the low amount of fluorescence, are remarkable. Because the solar-blind photomultiplier is insensitive to the exciting blue laser light, only the dark current of the detector (less than 30 pps) contributes to the background signal. Because of an integration gate time of 40 μsec (repetition rate 15 pps) the dark current causes a background signal of less than one count per minute and thus the dominant noise source is the signal itself.

The three-photon excitation of CO is shown in Fig. 2. The observed transitions are identified as the P-, Q-, and R-branch lines, with use of the single-photon absorption spectrum recorded by Simmons, Bass, and Tilford.[10] Except the $O(3)$ band head [in front of the $P(4)$ line], the O and S branch lines are too weak to be seen in the recording of Fig. 2. The N and T branches have not been observed so far.

For the recording of this spectrum laser light of 62-kW peak power was focused with a lens of 100-mm focal length to a beam waist of 10 μm. The observed fluorescence was maximum at CO pressures of about 2 Torr. The decrease at higher pressures is in agreement with the observations in two-photon excitation of CO and N_2 (in-

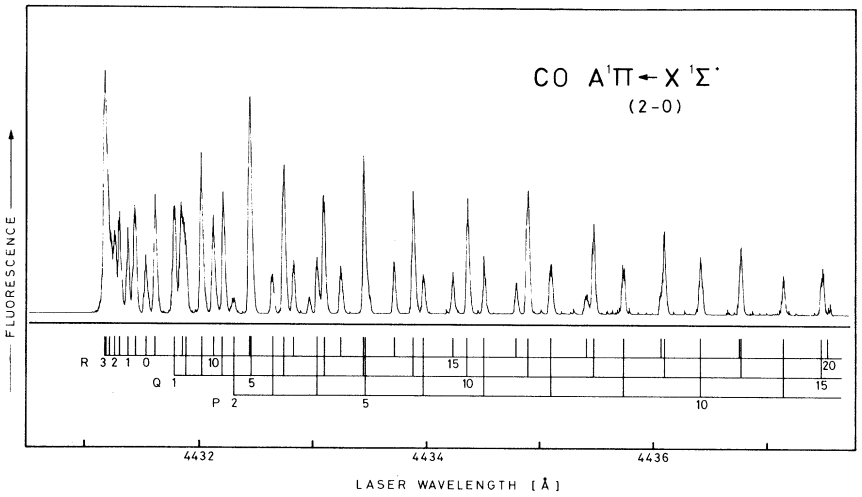

FIG. 2. Three-photon–excited fluorescence in CO recorded at a rate of 0.08 Å per minute and an effective time constant of $\tau = 3$ sec. CO pressure is 2 Torr.

volving mixtures[3] of CO and N_2) and are interpreted as due to quenching of CO ($A^1\Pi$, $v' = 2$) by ground-state CO. From the intensity of the strongest lines (Fig. 2) we estimate that about 10^4 excited molecules are produced per laser pulse. Because of the extremely low background signal, the three-photon resonances (Fig. 2) are recorded with good signal-to-noise ratio.

In nonlinear multiphoton spectroscopy ac Stark shifts and line broadening can severely limit the resolution and accuracy of the recorded spectra.[11] In two-photon excitation, it is usually possible to find experimental conditions for which ac Stark effects are sufficiently small. For the recording of three-photon absorption, however, considerably higher power densities are required. Thus light shifts and line broadening may be considerable. For the present three-photon resonance in Xe, for example, we observed a line broadening of up to 50 GHz at a power density of about 100 GW/cm^2.[12] From the linewidths measured at power densities between 4 and 100 GW/cm^2, we conclude that the resonance displayed in Fig. 1 is broadened by less than 10% of the observed width. This width of about 2.5 GHz results from Doppler broadening, the laser linewidth, and unresolved isotope shift.

In high-resolution three-photon spectroscopy the contributions of the ac Stark effect to measured line positions and linewidths have to be determined carefully and extrapolated to zero intensity. In time-resolved spectroscopy after pulsed three-photon excitation, however, these effects will be without any importance.

It should be noted that the Xe resonance will be observable at power densities considerably lower than 4 GW/cm^2. Using photon counting techniques the detector noise (with a discriminator bias of $\frac{1}{4}$ of the average single-electron pulse height) is about 2 counts per second. With a counting gate time of 100 nsec (repetition rate 15 pps) the background will be less than 1 count within 10^5 seconds. Therefore the signal-to-noise ratio will be limited only by the sampling time which will be limited ultimately by the operating time and frequency stability of the laser source.

From the measurements presented in this Letter we conclude that three-photon excitation should be a useful tool for high-resolution spectroscopy in atoms and molecules in the spectral region of the vacuum ultraviolet. With the powerful frequency-doubled tunable-laser radiation which can presently be generated at wavelengths down to 2170 Å, three-photon absorption will allow spectroscopic studies at wavelengths as short as 723 Å.

Besides the extension of nonlinear laser spectroscopy to such short wavelengths, the light of frequency-doubled lasers will permit doppler-free recording of three-photon absorption without doppler-broadened background. This can be achieved simply by simultaneous absorption of one ultraviolet photon and two visible photons of the fundamental laser frequency traveling in opposite direction to the ultraviolet photon.[13] Since many molecular states can also be excited by absorption of two photons of the ultraviolet light, a simultaneous measurement of Doppler-free two-

and three-photon transitions of the same molecular state becomes feasible. As in simultaneously recorded one- and two-photon spectra[14] of NO ($A^2\Sigma^+$) Doppler-free two- and three-photon absorption will provide a new method to obtain valuable spectroscopic information such as, for example, separations between two- and three-photon absorption lines which originate from λ-type doubling.

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Observation of Anomalous Resistivity Caused by Ion Acoustic Turbulence

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The dc resistivity caused by ion acoustic turbulence in plasmas is measured and effective collision frequencies of electrons are obtained from the resistivity as a function of electron drift velocity. These experimental results can be explained by using the theoretical model of Horton *et al.*, in which the nonlinear ion Landau damping dominates the turbulence spectrum.

Observation of anomalous electrical resistivity caused by ion acoustic turbulence in plasmas has been reported by many authors.[1-6] Collective electric fields associated with the ion acoustic instability[7] provide an effective high collision rate for electrons which, experimentally, appears as anomalously high electrical resistivity. Therefore, measurements of turbulence spectrum are very important for explaining the effective high collision rate since it would give more detailed information of the turbulence. However, the relationship between the effective collision frequency and the turbulence spectrum has not been investigated experimentally in detail so far. We wish to report here that the anomalous resistivity caused by the current-driven ion acoustic instability was observed in a large-diameter plasma and the dependence of the effective collision frequency on the electron drift velocity can be explained by using the theoretical results which have been derived by Horton and co-workers.[8,9]

The experiments were performed using the plasma box,[10] whose diameter and length were 70 cm and 34 cm, respectively. The plasma was produced by discharges between the tungsten filaments and the chamber wall with permanent magnets. The density and temperature of the electrons were measured by a Langmuir probe and