Laser-Induced Continuum Structure in Xenon

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Laser-induced continuum structure with a Fano-type line shape has been observed in xenon by the coupling of a bound $[5p^5 10p(J=0)]$ state to the ionization continuum with a tunable dye laser. The continuum was probed by three-photon ionization, two-photon–resonantly enhanced by the $5p^5 6p(J=0)$ state.

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Continuum structure similar to autoionizing resonances can be induced in atoms by the use of strong fields produced by intense lasers.[1] In this case, illustrated in Fig. 1, the interaction between the bound state $|k\rangle$ and the continuum $|\epsilon\rangle$ is electric dipole in nature, rather than electrostatic as in the case of normal autoionization, but the line shape, determined by probing from a populated state $|j\rangle$ with a weak field of frequency ω_p is Fano type, with a profile given by[2,3]

$$R(e)=(q+e)^2/(1+e^2),$$

where q is a dimensionless line-shape parameter and e is the frequency detuning from the Fano resonance, normalized with respect to its half width $\Gamma_{k\epsilon}$.

The position of the resonance is now determined by the energy of the bound state $|k\rangle$ and the frequency ω_d of the "dressing" radiation field, and by $\Gamma_{k\epsilon}$ which is given by

$$\Gamma_{k\epsilon}\simeq\pi|V_{k\epsilon}|^2.$$

$V_{k\epsilon}$ is the strength of the interaction which, for an electric dipole d is given by

$$V_{k\epsilon}=-\mathbf{d}_{k\epsilon}\cdot\mathbf{E}_d,$$

where $\mathbf{E}_d$ is the dressing electric field. Thus, the width of the Fano-type resonance is controlled by the intensity of the dressing laser.

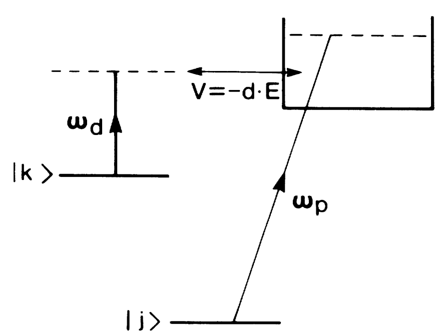

FIG. 1. Diagramatic representation of an atom, dressed by a photon ω_d and probed by ω_p.

In the case of the laser-induced continuum structure, the expression for the line-shape asymmetry parameter q is different from that developed in the general theory because of angular momentum selection rules for the dipole interactions. Application of these selection rules leads to an alternative interpretation of the q parameter in this case. It is found to be the ratio of the real to imaginary parts of the two-photon Rabi frequency for the interaction. The two-photon Rabi frequency has an imaginary part through the coupling of bound levels to the photoionization continuum. The continuum intermedium states are those which generate the usual induced continuum structure; it is, however, possible to couple the initial and final bound states through a Raman process via real bound states. These contribute only to the real part of the two-photon Rabi frequency and thus increase $|q|$.

Most previous experimental investigations of laser-induced continuum structure have used a weak probe interaction to study structure embedded in a continuum by a second, intense, dressing laser. These probe experiments test weak excitation of Fano resonances but do not provide information about the unusual saturation properties of intense-field excitation. We report here a study of the line shapes of induced resonances in strong fields. Feldmann *et al.*[4] have made a systematic investigation of resonances induced in the multiphoton ionization spectrum of sodium by a second strong laser field. They find in their experiments that a Raman coupling between the initial and "embedded" states through low-lying states dominates their ionization spectrum. These bound-bound Raman couplings increase the Fano q factor to such an extent that symmetric line profiles are generated[5,6]; these Raman processes obscure the induced continuum structure of interest here. In xenon, there are no low-lying states in the neighborhood of a Raman resonance, and the Raman coupling does not make a significant contribution; we would expect to observe asymmetric resonances associated with a small value of q.

Laser-induced continuum structure has been used to enhance third-harmonic generation in sodium vapor via a structured continuum[7] and to produce optical-polarization rotation in the neighborhood of an induced resonance.[8] The ability to control the strength of the photoionization continuum can lead to enhancement of nonlinear frequency-mixing processes by directly strengthening weak continua leading to an increase of coupling between the atom and the radiation field. However, by making use of the Fano minimum, it is possible, in principle, for us to weaken strong continua whose photoionization absorption would otherwise preclude the generation of useful intensities of vacuum radiation at frequencies above the ionization limit. Furthermore, the ability to suppress parasitic multiphoton ionization in, for example, resonant frequency mixing[9] may enable this saturation mechanism to be switched off. This permits the efficiency of the generation of radiation at frequencies below the ionization limit to be increased.

In this paper we report the observation of laser-induced continuum structure in xenon, observed by two-photon–resonant, three-photon ionization as shown in Fig. 2. The laser-induced continuum structure can be monitored by our keeping ω_1 fixed on the two-photon resonance and, by tuning of ω_d, the induced feature in the continuum can be scanned past the ionization energy $3\hbar\omega_1$. The alternative approach of keeping ω_d fixed and varying ω_1 introduces complications due to tuning through the two-photon–resonant state.

The condition that a bound state should be mixed with the continuum at energy $3\hbar\omega_1$ by a dressing field of photon energy ω_d is given by

$$3\hbar\omega_1 - \hbar\omega_d < E_{\text{I.P.}},$$

where $E_{\text{I.P.}}$ is the ionization potential of the atom. If in addition $2\hbar\omega_1 + \hbar\omega_d > E_{\text{I.P.}}$, nondegenerate two-photon–resonant three-photon ionization is possible and irradiation of the atom by both optical fields will produce a net increase in the total ionization, compared with that obtained with only ω_1 present, i.e., if $E_{\text{I.P.}} < \frac{5}{2}\hbar\omega_1$, the induced continuum structure will be superimposed on an enhanced background due to $2\omega_1 + \omega_d$. While the use of noble gases such as xenon offers considerable experimental advantages over atomic vapors, the fact that all the two-photon–resonant states are high lying results in this increase of the total background ionization signal. Under these conditions, saturation of two-photon–resonant frequency-mixing processes due to ionization broadening of the two-photon state cannot be substantially decreased.

The experimental system comprised two excimer-pumped (XeCl) tunable, narrow-bandwidth dye lasers, one of which was frequency doubled to 249.63 nm, the wavelength required to achieve two-photon resonance with the $[5p^6\text{-}5p^56p(J=0)]$ transition, and then amplified in a single-pass KrF amplifier. The $5p^510p(J=0)$ state was mixed with the continuum in the region of the three-photon energy $(3\hbar\omega_1)$ with the second laser tuned to $\lambda_d \simeq 401.73$ nm, and the ionization signal was monitored in an ionization chamber. The experimental arrangement is shown in schematic form in Fig. 3.

Both dye lasers were line narrowed and tuned with both a grazing-incidence grating and a second Littrow grating in place of the more usual mirror.[10] When they were pumped close to threshold, the output was observed to be in a single longitudinal mode, with an estimated bandwidth of 100 MHz, although at higher pumping powers a second mode was produced giving a bandwidth of 1 GHz. The output pulse form of both lasers was approximately Gaussian with a pulse width of 12 ns. The pulse from the probe laser (coumarin 307 in ethanol) was amplified in a two-stage amplifier to 2 mJ; the second-harmonic radiation at 249.63 nm was generated with a crystal of potassium pentaborate and amplified to 0.5 mJ in a single-pass KrF amplifier (Lambda Physik Model 101). The dressing laser pulse {PBBO [2-(4-biphenylyl)-6-phenylbenzoxazol-1,3] in dioxan} was also amplified in a two-stage amplifier, the second amplifier containing DPS (diphenylstilben) in dioxan. The output energy was 1.5 mJ. The amplified pulse was passed through a defocused 1:1 telescope (T) before being combined with the probe laser at a dichroic mirror. The defocused telescope permitted variation of the relative positions of the foci of the two beams produced by the singlet planoconvex lens (L) used to focus into the ionization

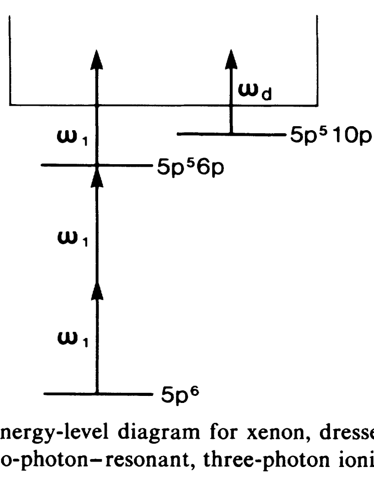

FIG. 2. Energy-level diagram for xenon, dressed by ω_d and probed by two-photon–resonant, three-photon ionization.

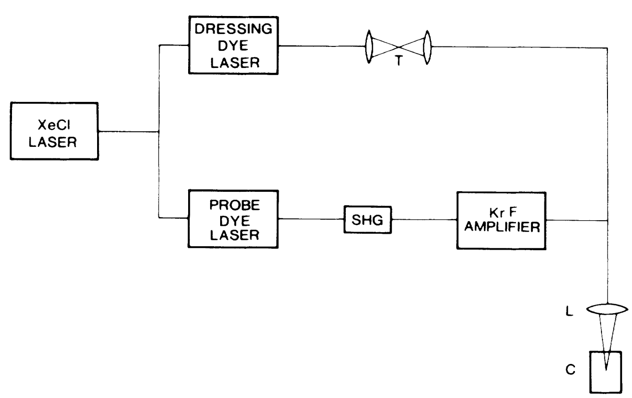

FIG. 3. Schematic diagram of laser system.

chamber (C). The ionization chamber contained xenon at a pressure of 5–10 Torr, and had guard rings to ensure that ionization was measured only from a specified volume where the overlap of the beams could be carefully controlled.

The frequency of the probe laser (ω_1) was tuned to the two-photon resonance in xenon by monitoring of the two-photon–resonant, three-photon ionization signal with the dressing-laser beam blocked. In order to observe continuum structure with the dressing laser present it is necessary to ensure that the multiphoton ionization is not saturated because of depletion of ground-state atoms. From a study of the saturation characteristics of the ionization with only the probe laser, a working range of 10–100 MW cm^{-2} was identified for experiments with the dressed atoms.

Figure 4 shows a plot of the total two-photon–resonant three-photon ionization signal versus ω_d, the frequency of the dressing laser. There is clear evidence of Fano-type structure. The absence of a precise zero in the ionization signal is due in part to nondegenerate three-photon ionization ($2\omega_1+\omega_d$) discussed earlier. However, the width of the Fano profile depends on the dipole interaction potential $V_{k\epsilon}$ and hence the intensity of the dressing laser. For a pulsed laser with a time-varying intensity, the spectral profile of the measured temporally integrated ionization signal is modified so as to reduce both the height of the peak and depth of the minimum of the conventional Fano profile. Furthermore, it has been shown[11,12] that, for strong probing fields, the distinction between probing and dressing fields becomes artificial and both lasers can be considered to dress the atom. In this case the induced structure becomes time dependent even for constant-amplitude fields. For pulsed fields, the interference structure responsible for the Fano-type resonance becomes time dependent. An analysis of structure induced by strong dressing fields is to be published.[13]

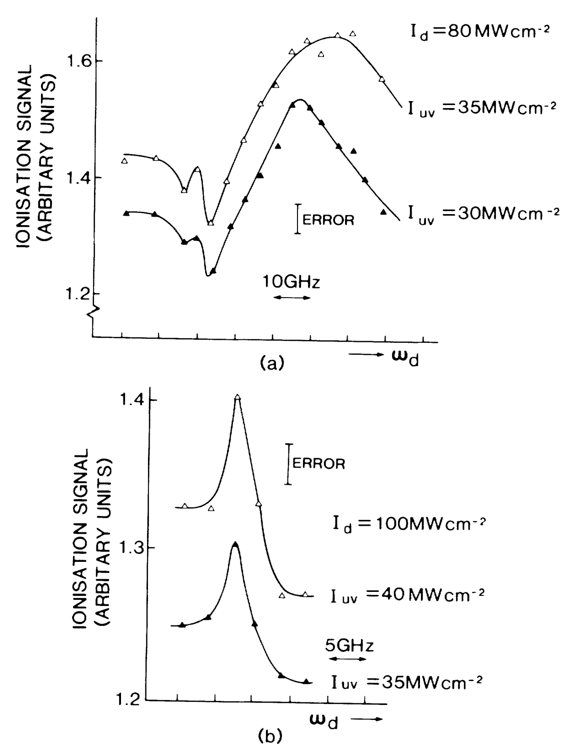

FIG. 4. Plots of the total two-photon–resonant ionization signal vs ω_d: (a) Low-resolution scan and (b) higher-resolution scan of the unknown feature.

An artificially high background can, in principle, arise from nonresonant five-photon ionization at frequency ω_d and incomplete temporal and spatial overlap of the two laser pulses, but these effects were found to be insignificant.

There is evidence of a secondary peak on the low-frequency side of the principal Fano minimum. A higher-resolution scan of this region with use of slightly higher uv and dressing-laser intensities is shown in Fig. 4(b) and this indicates a peak of approximately 5-GHz width. This feature was observed consistently over a range of probing and dressing laser intensities and is not believed to be a systematic error. However, the origin of this feature is not known.

In conclusion, laser-induced continuum structure has been observed in xenon with use of intense pulses. This process may have general applications in controlling multiphoton interactions in atoms above the ionization limit.

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