PHYSICAL REVIEW LETTERS

VOLUME 33 | 11 NOVEMBER 1974 | NUMBER 20

Autoionizing States of Sr Studied by the Generation of Tunable Vacuum uv Radiation

J. A. Armstrong and J. J. Wynne

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

(Received 24 September 1974)

We analyze the spectral profile for the resonant enhancement of four-wave parametric generation in the vicinity of autoionizing transitions. A comparison between theory and experiment is presented for an autoionizing line of Sr I seen at 1867 Å.

Hodgson, Sorokin, and Wynne[1] have reported the generation of tunable vacuum uv (vuv) light by means of resonantly enhanced[2] four-wave parametric interactions in Sr vapor. In certain wavelength ranges it is observed that the spectrum of the *generated* vuv radiation is dominated by autoionizing states of Sr. These states have heretofore been seen only in optical absorption or in electron energy-loss spectra,[3,4] for which cases the lines have the so-called Fano-resonance shapes.[4] We report here the comparison of experimental and theoretical spectral profiles for an autoionizing line of Sr I seen at 1867 Å by means of four-wave parametric generation.[5]

The experiments were carried out with two dye lasers, of frequencies ν_1 and ν_2, the generated light having frequency $\nu_{\text{vuv}}=2\nu_1+\nu_2$. One laser is tuned so that $2h\nu_1$ equals the energy of a two-photon allowed transition from the ground state. This results in a large enhancement of the nonlinear susceptibility. The second laser is then tuned, scanning ν_{vuv} through the autoionizing level, with a resulting rapid variation of the intensity of the generated signal as a function of frequency. Because of the resonant enhancement, the nonlinear susceptibility,[6] $\chi^{(3)}$, may be approximated by

$$\chi^{(3)}(\nu) \propto \sum_{k \text{ only}} P_{gk}P_{kj'}P_{j'j}P_{jg}/(\nu_{kg}-\nu_{\text{vuv}})(\nu_{j'g}-2\nu_1)(\nu_{jg}-\nu_1). \tag{1}$$

The ground state in Eq. (1) is labeled $|g\rangle$, j and j' label discrete, excited states, and k labels the continuum—including the autoionizing state—which is connected both to $|g\rangle$ and to $|j'\rangle$ by the electric dipole matrix elements P_{gk} and $P_{kj'}$. For the case of Sr I, state $|g\rangle$ is $5s^2\,{}^1S_0$, the particular autoionizing state at 1867 Å reported on in this paper belongs to the $4d4f$ configuration,[7] and the two-photon intermediate state $|j'\rangle$ is chosen to be either $5p^2\,{}^1D_2$ or $5s5d\,{}^1D_2$.

According to Fano[4] each matrix element in (1) involving the continuum may be put in the form

$$P_{kj'}=(\pi V_E{}^*)^{-1}\langle\Phi|P|j'\rangle\sin\Delta-\langle\psi_E|P|j'\rangle\cos\Delta, \tag{2}$$

where ψ_E is the unmodified continuum state of energy E, and Φ is the modified, discrete part of the autoionizing state, which has been shifted and broadened in energy by its interaction with the continuum. V_E is the configuration-interaction matrix element between ψ_E and the unmodified discrete state φ. The only quantity in Eq. (2) which varies rapidly is Δ, which is defined to be $\Delta=\arctan[\pi|V_E|^2/(\bar{E}-E)]$, where $\bar{E}$ is the energy of the state Φ.

Under appropriate experimental conditions the generated light will have the spectrum of $[\chi^{(3)}(\nu)]^2$

alone, which we find to have the following frequency dependence:

$$[\chi^{(3)}(\nu)]^2 \propto (1+x^2)^{-2}[q_g + q_{j'} + x(q_g q_{j'} - 1)]^2 (P_{g\psi_E} P_{\psi_E j'})^2. \tag{3}$$

Here $x \equiv (h\nu_{\rm vuv} - \bar{E})/\pi |V_E|^2$. The parameter $q_j \equiv P_{\Phi j}/\pi V_E{}^* P_{\psi_E j}$ was introduced by Fano to characterize the shape of an absorption line due to a transition between the discrete level j and the autoionizing level of interest.

The result given in Eq. (3) was obtained from a calculation whose outline is as follows: Because $2\nu_1$ is fixed and chosen to resonate with a particular two-photon state, the only significant frequency dependence of $\chi^{(3)}$ comes from the sum over k of the first two matrix elements in the numerator of Eq. (1) divided by the first resonant denominator. With the use of the appropriate version of Eq. (2) for both P_{gk} and $P_{kj'}$, and the realization that both expressions involve the same Δ (since Δ is a property of the autoionizing state alone), it is simple to do the integration required in Eq. (1).[8] The result is given in Eq. (3), which applies when the autoionizing resonance is many linewidths removed both from ionization limits and from other autoionizing levels. [This enables the integrals in (1) to be extended effectively from $+\infty$ to $-\infty$. If other states or ionization limits are close by, the expression for $\chi^{(3)}$ may still be worked out easily, but is more complicated than Eq. (3).]

When q_g is known for the transition between the ground state and a particular autoionizing level, Eq. (3) and the observed vuv parametric-generation spectrum allow one to infer the $q_{j'}$ for the transition between an excited discrete state, j', and the given autoionizing level. This information is not readily obtainable by other means. Several calculated line shapes are shown in Fig. 1. Here, q_g is set equal to 1 and the graph shows how the line shape depends on $q_{j'}$. In particular, note that the curves are symmetric in $h\nu_{\rm vuv} - \bar{E}$ for $q_{j'} = \pm 1/q_g = \pm 1$, as expected from inspection of Eq. (3). Any other value of $q_{j'}$ produces an asymmetric curve, with a zero at $x = (q_g + q_{j'})/(1 - q_g q_{j'})$.

Experimentally, we studied the generation near 1867 Å involving the $4d4f$ level under two conditions: First, the two-photon intermediate state $|j'\rangle$ was chosen to be the $^1D_2$ arising from the $5s5d$ configuration. The generated light had a symmetric line shape [Fig. 2(a)] which could be well matched by Eq. (3) for $q_g = 3.5$ (known from Ref. 7) and $q_{j'} = 1/3.5 \simeq 0.29$. From this fit we deduce a linewidth of 25 cm^{-1}, in agreement with the value of 20.3 cm^{-1} given[9] in Ref. 7. In the second case, the first laser was tuned so that $2\nu_1$ resonated the $^1D_2$ state arising from the $5p^2$ configuration. The resulting generation at 1867 Å in this case is strongly asymmetric [Fig. 2(b)] but may be fairly well fitted by Eq. (3) with $q_g = 3.5$ (as before), $q_{j'} \simeq -0.5$, and a linewidth of 25

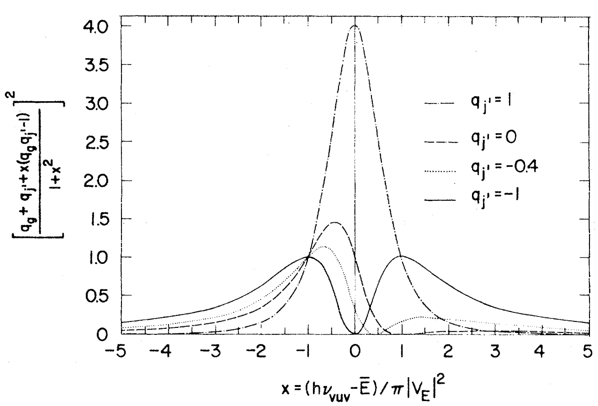

FIG. 1. Variation of $|\chi^{(3)}|^2$ as a function of $\nu_{\rm vuv}$ for $q_g = 1$ and for several values of $q_{j'}$.

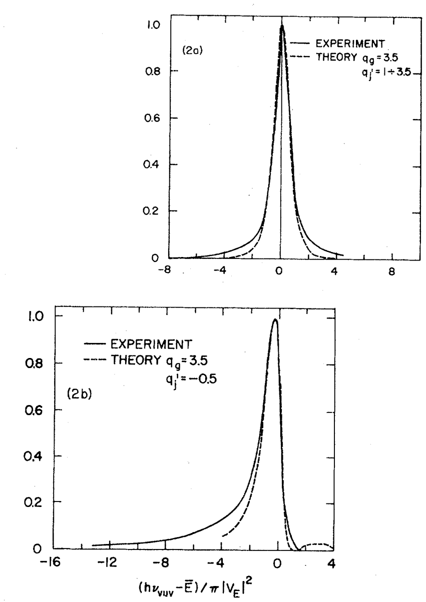

FIG. 2. Comparison of experiment and theory for the line shape of vuv generation near 1867 Å. In (a) the intermediate state is $5s5d\,^1D_2$, and in (b) the intermediate state is $5p^2\,^1L_2$. The vertical scales are arbitrarily set to unity at the maximum.

cm^{-1}. The line shape allows one to determine the approximate magnitude and the sign of $q_{j'} = P_{\Phi j'}/\pi V_E^* P_{\psi_E j'}$ for two different intermediate states, j'. We do not yet understand why the experimental shapes have higher wings than the theoretical.

Thus, we have a new technique for studying autoionizing levels. By determining q for transitions between an autoionizing level and several intermediate states (j'), one can gain information to supplement what is known about the level from absorption spectroscopy. This can lead to new classifications for the quantum numbers of the discrete-state component of the autoionizing level. An accurate relative measurement of $|\chi^{(3)}|^2$ using different j' levels determines the $q_{j'}$ and the ratios of the $P_{\Phi j'}$. To do this accurately requires a knowledge of the coherence length for the parametric mixing process,[1] the absorption coefficients at ν_1, ν_2, and ν_{vuv} as functions of frequency, and a measurement of the intensities of the light at ν_1, ν_2, and ν_{vuv}. For our measurements in Sr, the weak oscillator strength of the autoionizing transition and its broad linewidth resulted in negligible contributions to the coherence length and absorption coefficient, so that the observed line shape was characteristic of $|\chi^{(3)}(\nu)|^2$ alone, but we did not measure the relative intensities.

We gratefully acknowledge extensive discussions with Dr. P. P. Sorokin and Dr. R. T. Hodgson and the technical assistance of L. H. Manganaro.

[1]R. T. Hodgson, P. P. Sorokin, and J. J. Wynne, Phys. Rev. Lett. 32, 343 (1974).

[2]R. B. Miles and S. E. Harris, IEEE J. Quantum Electron. 9, 470 (1973); A. H. Kung, J. F. Young, G. C. Bjorklund, and S. E. Harris, Phys. Rev. Lett. 29, 985 (1972).

[3]G. V. Marr, *Photoionization Processes in Gases* (Academic, New York, 1967), Chap. 7.

[4]U. Fano, Phys. Rev. 124, 1866 (1961).

[5]A preliminary report of this work was given as a post-deadline paper (P-9) at the Eighth International Quantum Electronics Conference, San Francisco, California, 10–13 June 1974.

[6]J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).

[7]W. R. S. Garton, G. L. Grasdalen, W. H. Parkinson, and E. M. Reeves, J. Phys. B: Proc. Phys. Soc., London 1, 114 (1968).

[8]E. C. Kemble, *The Fundamental Principles of Quantum Mechanics* (McGraw-Hill, New York, 1937), Chap. XI.

[9]The values of Γ given in Ref. 7 are in units of circular frequency and must be divided by $2\pi c$ to give wave numbers.

Low-Temperature Studies of the Rayleigh-Bénard Instability and Turbulence

Guenter Ahlers
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 20 August 1974)

Low-temperature techniques were applied to the study of a hydrodynamic instability. High-precision results for the Nusselt number N as a function of the Rayleigh number R for liquid and gaseous helium revealed no singularities in $N(R)$, except at the convective threshold R_c. For $R > 2.19R_c$, a new turbulent state was found and characterized by measuring the frequency spectrum and the amplitude of $N(R)$.

I wish to report on a number of quantitative experimental results relating to heat transport by thermal convection at low temperatures in liquid and gaseous He^4. The measurements were made on a horizontal layer of the fluid heated from below. They thus pertain to the Rayleigh-Bénard instability,[1] a particularly simple case of a hydrodynamic instability which has caused considerable interest[2-6] among physicists recently. The present work exploits some of the experimental advantages of low-temperature techniques[7] which permit thermal measurements of very high resolution and great accuracy. In addition to providing accurate measurements of the onset of convection and of the heat transport by the fluid under a wide range of conditions, the experiments reveal a transition to, and provide a quantitative description of, a new turbulent state. The properties of this state are described rather well by a theory developed recently by McLaughlin and Martin.[6]

The apparatus has been described in detail else-