COMMENTS

Comment Concerning the Study of Autoionizing States Using Parametric Generation*

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A recent experiment utilizing four-wave parametric generation to study autoionizing states has been reanalyzed, using a different theoretical line shape from that proposed in the original article. The agreement between theory and experiment is found to be excellent.

In a recent Letter,[1] Armstrong and Wynne have demonstrated the feasibility of using parametrically generated, tunable, vacuum-ultraviolet radiation (vuv) to study autoionizing states of atomic systems. Using the Fano formalism,[2] the authors derive an expression for the line shape of the vuv as a function of the frequency of the incident light, and use this expression to fit their experimental data for Sr I. From these fits, they are able to obtain widths and q values (as defined by Fano[2]) for the autoionizing states.

As remarked by Armstrong and Wynne, the experimental line shapes obtained are noticeably higher in the wings than are the corresponding theoretical line shapes. Thinking that this interesting discrepancy might be caused by power-broadening effects, we undertook a calculation of the process using the resolvent formalism.[3] Taking the low-intensity limit of our calculation, we found that our result disagreed significantly with the expression for the vuv line shape [Eq. (3) of Ref. 1] given by Armstrong and Wynne.

Because the theoretical line shape is essential to the proper interpretation of the experimental data, we attempted to resolve this discrepancy. It seems to be related directly to the method used by Armstrong and Wynne to evaluate the integrals involved in the calculation.

In performing the integral required to go from Eq. (2) to Eq. (3) of Ref. 1, Armstrong and Wynne interpreted the integral as a principle-part integral.[4] However, in general, when a perturbation calculation involves intermediate states which belong to a continuum, the resulting integrals are not taken totally along the real axis, but are deformed so as to pass below all poles which appear on the real axis.[5] This corresponds to replacing the resonant denominator $(\nu_{kg}-\nu_{\text{vuv}})^{-1}$ of Eq. (2), Ref. 1, by $\text{P}(\nu_{kg}-\nu_{\text{vuv}})^{-1}+i\pi\delta(\nu_{kg}-\nu_{\text{vuv}})$. The result obtained by using this expression agrees with results obtained using a resolvent formalism.

The expression obtained in this way for the nonlinear susceptibility $|\chi(x)|^2$ is given by

$$|\chi(x)|^2 \propto \frac{(q_g q_{j'})^2+(q_g+q_{j'}+x)^2}{1+x^2}, \qquad (1)$$

where q_g and $q_{j'}$ are the Fano parameters[2] for the states $|g)$ and $|j')$, respectively, and x is the normalized energy[1] $(\hbar\nu_{\text{vuv}}-\bar{E})/\frac{1}{2}\Gamma$, with $\bar{E}$ the en-

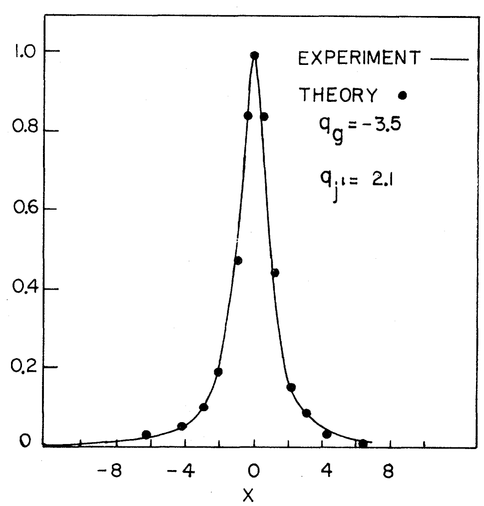

FIG. 1. Comparison of experimental data of Armstrong and Wynne (Ref. 1) to theory [Eq. (1)] for the intermediate state $5s5d\,^1D_2$. The vertical scale is arbitrarily set to unity, and $x=(\hbar\nu_{\text{vuv}}-\bar{E})/\frac{1}{2}\Gamma$.

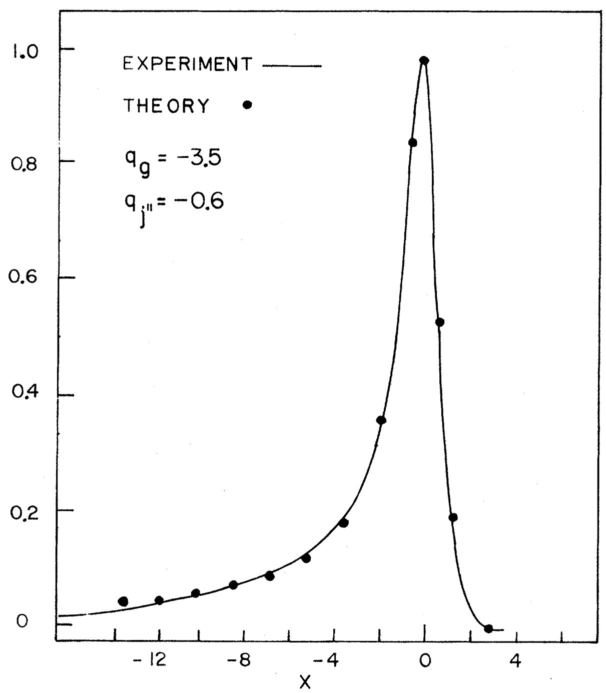

FIG. 2. Comparison of experimental data of Armstrong and Wynne (Ref. 1) to theory [Eq. (1)] for the intermediate state $5p^2\,{}^1D_2$. The vertical scale is arbitrarily set to unity, and $x = (\hbar\nu_{\mathrm{VUV}} - \bar{E})/\frac{1}{2}\Gamma$.

ergy separation between the autoionizing state φ and $|j')$ and $\Gamma = 2\pi |V_E|^2$ the linewidth of the autoionizing state. It is this expression which should be utilized in the interpretation of data obtained in this type of experiment.

We have used Eq. (1) to analyze the data of Armstrong and Wynne.[4] There are five parameters involved in the analysis of each curve: q_g, $q_{j'}$, the width Γ, a scaling factor which turns Eq. (1) into an equality rather than a proportionality, and the value of the background signal upon which the experimental peaks are superimposed. When all five parameters were allowed to vary freely, only Γ was determined reasonably well. As it was obviously necessary to decrease the number of variables, we fixed q_g at the value obtained by Garton *et al.*,[6] $q_g = -3.5$; a subsequent fitting of the data then produced unambiguous values of the remaining four parameters. The data for the $|j') = |5s5d\,{}^1D_2)$ intermediate state resulted in the values $\Gamma = 16.5$ cm^{-1} and $q_{j'} = 2.1$. It was noted, however, that these data are relatively insensitive to the values of q_g and $q_{j'}$. Figure 1 shows the excellent agreement between theory and experiment in this case. The data for the $|j'') = |5p^2\,{}^1D_2)$ intermediate state result in an almost identical value of the common parameter Γ, $\Gamma = 17.5$ cm^{-1}, and a very small value for $q_{j''}$, $q_{j''} = -0.6$. The comparison between experiment and theory in this case is shown in Fig. 2. The values quoted above for Γ are essentially identical to those obtained when all five parameters were allowed to vary and are not inconsistent with the value for Γ found by Garton *et al.*,[6] of 20.2 cm^{-1}.

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[1]J. A. Armstrong and J. J. Wynne, Phys. Rev. Lett. 33, 1183 (1974).

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

[3]See, e.g., W. Heitler, *The Quantum Theory of Radiation* (Clarendon, Oxford, England, 1960); M. L. Goldberger and K. M. Watson, *Collision Theory* (Wiley, New York, 1964).

[4]J. A. Armstrong, private communication.

[5]D. R. Bates, in *Quantum Theory,* edited by D. R. Bates (Academic, New York, 1961), Vol. I, p. 282. For a more advanced discussion, see Ref. 3.

[6]W. R. S. Garton, G. L. Grasdalen, W. H. Parkinson, and E. M. Reeves, J. Phys. B: Proc. Phys. Soc., London 1, 114 (1968). We note that the energy parameter used by these authors is the negative of that used by Fano; this forces their q's to be the negative of the Fano definition, which we follow.