Collisionally Induced Narrowing of the Longitudinal Relaxation Linewidth in Nearly Degenerate Four-Wave Mixing

J. F. Lam, D. G. Steel, and R. A. McFarlane
Hughes Research Laboratories, Malibu, California 90265
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This paper reports the observation of collisionally induced spectral narrowing of the longitudinal relaxation linewidth with use of the nearly degenerate four-wave mixing process. A theoretical model, which takes into account strong velocity-changing collisions for the population and phase-interrupting collisions for the optical coherence, leads to a good qualitative understanding of this new effect.

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This communication describes the first observation of a collisonal narrowing of the longitudinal relaxation linewidth which we observe using nearly degenerate four-wave mixing (NDFWM). The origin of the effect is explained by an analytical treatment of NDFWM which includes spontaneous emission and the effects of collisions due to ground-state perturbers. The results show that a direct measurement of velocity-changing collision rates may be possible with this method.

We consider the nonlinear interaction between three input fields denoted by $\vec{E}_f$, $\vec{E}_b$, and $\vec{E}_p$ oscillating at frequencies ω, ω, and $\omega+\delta$, respectively, and a set of moving two-level atoms having transition frequency ω_0. $\vec{E}_f$ and $\vec{E}_b$ are counterpropagating pump fields and $\vec{E}_p$ is the probe field which is nearly collinear to $\vec{E}_f$.[1] The physical process giving rise to a four-wave mixing signal in such a system can be described as follows. The interference of the fields $\vec{E}_f$ and $\vec{E}_p$ generates a nonequilibrium population difference which propagates in space with phase velocity $\delta/|\vec{K}_f-\vec{K}_p|$. The population difference provides a moving grating from which the field $\vec{E}_b$ scatters, generating a signal wave propagating in the opposite direction to $\vec{E}_p$. Conservation of energy imposes the condition that the frequency of the signal field is given by $\omega-\delta$. As a result of Doppler broadening, two resonances are observed, corresponding to two distinct velocity groups: The first occurs at $\delta=0$ when the forward pump and probe are resonant with one velocity group of atoms, and the second occurs at $\delta=-2\Delta$ ($\Delta=\omega_0-\omega$) when the forward pump and backward propagating signal are resonant with the second velocity group of atoms. The linewidths of the two resonances are given by the energy relaxation time (T_1) and the dipole dephasing time (T_2), respectively.[2,3]

Because of the distinct nature of the processes determining the bandwidths of the resonances at $\delta=0$ and $\delta=-2\Delta$, collisions due to ground-state perturbers affect the linewidths of the two resonances differently. The bandwidth associated with the dephasing of the optical coherence in the presence of buffer gases is affected by phase-interrupting collisions and gives rise to collisional broadening of the $\delta=-2\Delta$ resonance.[4] The bandwidth associated with the population relaxation in the presence of buffer gases is a manifestation of velocity-changing collisional processes in addition to radiative decay processes. This paper shows that these effects give rise to collisional narrowing of the $\delta=0$ resonance. In principle, NDFWM permits a simultaneous examination of collisional dynamics for each of these processes, leading to independent measurements of velocity-changing and phase-interrupting pressure-dependent cross sections.

The origin of collisional narrowing of the longitudinal (T_1) relaxation linewidth arises from the physical decoupling of the ground and excited states in the presence of buffer gases. In the absence of foreign perturbers, the ground and excited states are coupled together by means of the vacuum radiation field (the strength of this coupling is measured by the spontaneous decay rate γ). As a result of this coupling, both quantum states evolve as a single entity even in the presence of applied radiation fields, and this entity has a unique spectral response, i.e., the bandwidth is determined by γ. In the presence of foreign perturbers, the ground and excited states experience different collisional interactions which effectively decouples them even in the presence of the vacuum radiation field. In this case, the responses of the ground and excited states to applied radiation fields reflect their evolution as distinct entities. Hence, the NDFWM signal has contributions arising from both the ground and excited states separately. The spectral response of the individual contribution is characterized by the respective collisional cross section. For the signal arising from the ground state, the bandwidth is determined by $\gamma_t+\Gamma_1$ while that of the excited state is determined by $\gamma+\gamma_t+\Gamma_2$. Γ_n is the velocity-changing collisional decay rate and γ_t is the reciprocal of the transit time. For low pressures, one notes that $\gamma_t+\Gamma_1 \ll \gamma+\gamma_t+\Gamma_2$. Hence, the contribution from the ground state will dominate over that from the excited state. The bandwidth is determined by $\gamma_t+\Gamma_1$ which is much narrower than γ. A measurement of the bandwidth at the resonance line $\delta=0$ will provide a direct measurement of the ground-state decay rate Γ_1 provided that the transit time is known accurately.

To test this physical picture we have calculated the effect of collisions on the spectral response of NDFWM, in the impact regime. We have chosen a collision regime which entails a complete thermalization of the velocity distribution upon each scattering event, i.e., the strong-collision model. The third-order optical polarization obtained with a perturbation solution of the density-matrix equations is given by

$$P(\vec{r},t)=-N_0\frac{|\mu_{21}|^4}{(2i\hbar)^3}E_fE_bE_p{}^*\{\exp[-i\vec{K}_p\cdot\vec{r}-i(\omega-\delta)t]\}\sum_{n=1}^{2}\{S_n+R_n\}, \quad (1)$$

where N_0 is the initial population of the ground state and μ_{21} is the dipole moment between the ground and excited states. In the extreme Doppler limit, S_n and R_n are given by

$$S_n=\left\{1+\frac{\gamma}{\gamma+\Gamma_2-\Gamma_1}(-1)^n\right\}I_n+\frac{\gamma}{\gamma+\Gamma_2-\Gamma_1}(-1)^nN_2\left\{\frac{\gamma+\Gamma_2+i\delta}{\gamma n+i\delta}\right\}, \quad (2a)$$

$$R_1=N_1,\quad R_2=N_2, \quad (2b)$$

$$I_n=-2\pi^{1/2}(Ku_0)^{-1}(\gamma_n+i\delta)^{-1}(2\gamma_e+i[2\Delta+\delta])^{-1} \quad (2c)$$

$$N_n=\frac{\Gamma_n}{Ku_0}\left(\frac{6\pi}{Ku_0}\right)\left(\frac{1}{\gamma_n+i\delta}\right)\left(\frac{1}{\Gamma_n+i\delta}\right), \quad (2d)$$

where $\gamma_e=\gamma/2+\Gamma_{12}$, $\gamma_1=\Gamma_1$, and $\gamma_2=\gamma+\Gamma_2$. Γ_{12} is the pressure-dependent dephasing rate and Ku_0 is the Doppler width. We assume that $\delta\ll\omega$ and transit-time effects have been included phenomenologically.

The quantities S_n and R_n have simple physical interpretations. S_n has two contributions. The first one, proportional to I_n, arises from the dynamics that gives rise to the resonances at $\delta=0$ and $\delta=-2\Delta$ with their respective linewidths modified by collisions. The second contribution, proportional to γ, is due to the existence of spontaneous decay from the velocity-redistributed excited state to the ground state. R_n, proportional to Γ_n/Ku_0 as shown in Eq. (2d), is due to velocity redistribution of the population in the presence of buffer gases. In the absence of buffer gases (turn off Γ_1, Γ_2, and Γ_{12}) the spectral response is given by the term proportional to I_n in Eq. (2a).

The calculated spectral response for various buffer-gas pressures is shown in Fig. 1. As buffer gas is added, we find that the amplitude at $\delta=-2\Delta$ decreases as a function of pressure, while the linewidth of that resonance broadens in accordance with the phase-interrupting collision model. However, we observe that the linewidth of the resonance at $\delta=0$ narrows as a function of pressure and the magnitude of the signal increases. The narrowing yields a minimum width comparable to the value determined by transit-time effects (assumed to be 1 MHz in this calculation). An increase in magnitude of the response which was found at $\delta=0$ arises from the tendency of collision processes to fill up the velocity hole "burned" by the radiation fields. (Magnitude changes are not indicated in the figures.)

As anticipated above, the narrowing of the linewidth at the resonance $\delta=0$ in the presence of buffer gas is due to the appearance of distinct quantum mechanical scattering amplitudes for the excited and ground states. The distinct scattering amplitudes translate into different cross sections resulting in different decay rates Γ_n. Hence, collision effects lead to distinct bandwidths for both states.

The experimental configuration for these collision studies was identical to that used by Steel and Lind.[5] The counterpropagating pump fields were supplied by one stabilized cw dye laser (CR699-21) while the nearly collinear probe wave was supplied by a second stabilized cw dye laser (also CR699-21). The lasers were tuned to the $3s\,^2S_{1/2}(F=2)$-$3p\,^2P_{3/2}(F=3)$ transition of the D_2 line in sodium at 589 nm. Beam intensities were on the order of 2 mW/cm^2. All three input beams

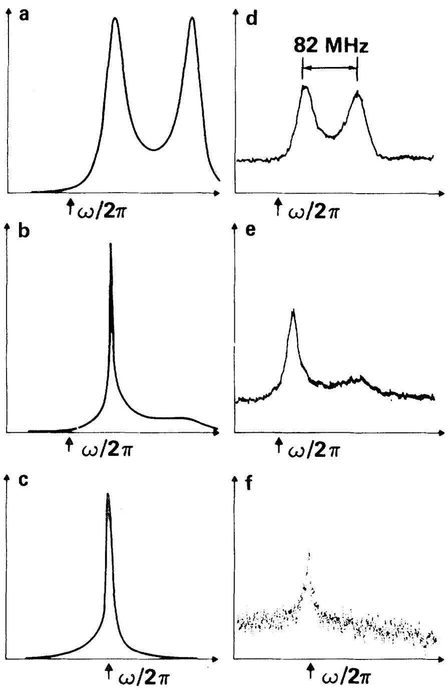

FIG. 1. Comparison between theory and experiment of the NDFWM spectral response for various neon buffer-gas pressures. The amplitude scaling varies with each figure. The separation between the double peaks is 82 MHz. (a) Theory, $\Delta = -41$ MHz, 0 Torr. (b) Theory, $\Delta = -41$ MHz, 2 Torr. (c) Theory, $\Delta = 0$, 10 Torr. (d) Experiment, $\Delta = -41$ MHz, 0 Torr. (e) Experiment, $\Delta = -41$ MHz, 2 Torr neon. (f) Experiment, $\Delta = 0$, 33 Torr neon.

were copolarized to avoid Zeeman coherence effects which are discussed elsewhere.[6]

Shown in Fig. 1(d) is a typical spectral response as the probe frequency is tuned near the pump frequency. These data are similar to those measured earlier.[5] In these experiments, it was important to tune the pump laser to the high-frequency side of the transition to avoid confusion with neighboring hyperfine levels in the upper state. Two resonances are observed occurring at $\delta = 0$ and at $\delta = -2\Delta$ as predicted in Fig. 1(a). As expected from the known lifetime of the transition, the widths of each peak were found to be approximately equal to 20 MHz. The width of the first peak is given by $1/\pi T_1$ while the width of the second peak is given by $2/\pi T_2$. The widths of the resonances are the same since we are using a ground-state transition for which $T_2 = 2T_1$ in the absence of collisions. In Fig. 1(e) we see the effect of adding a small amount (2 Torr) of neon buffer gas. As in the theoretical calculation of Fig. 1(b), there is a slight narrowing of the first peak at $\delta = 0$. The second peak has dropped in magnitude and broadened with respect to the first peak due to phase-interrupting collisions. The measured broadening of that peak is in agreement with the pressure-broadening rates tabulated by Lewis.[7] As the buffer-gas pressure is increased beyond a few Torr to the 30-Torr range, we see in Fig. 1(f) that the first peak is narrowed considerably. For these data, the pump frequency was adjusted to be coincident with the atomic resonance ($\Delta = 0$). However, similar narrowing behavior was observed when the pump frequency was tuned to the high-frequency side of the resonance, though the data were considerably noisier because of a reduced signal level. The narrowest linewidth measured was approximately 5 MHz but was limited by relative laser jitter.

The quantitative differences as a function of pressure between theory and experiments are most likely due to the collision model which assumes strong velocity-changing collisions. A more general description of collisions is presently being incorporated into the formalism. On the basis of this improved analysis and the above experiments we expect that by reducing the relative laser jitter and transit-time broadening, it should be possible to directly observe linewidths determined by velocity-changing collisions.

In conclusion, we have shown that in the presence of ground-state perturbers, the spectral response of the NDFWM signal shows a narrowing of the bandwidth of the longitudinal relaxation resonance ($\delta = 0$). We have presented a theory and a physical picture which qualitatively account for the observations showing the narrowing to be due to a collision-induced distinction between ground- and excited-state dynamics. These results demonstrate the possibility of determining the ground-state velocity-changing collision cross sections directly from the spectral response of NDFWM.

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Influence of Increasing Nuclear Charge on the Rydberg Spectra of Xe, Cs^+, and Ba^{++}: Correlation, Term Dependence, and Autoionization

W. T. Hill, III
National Bureau of Standards, Washington, D.C. 20234, and Institute for Physical Science and Technology, University of Maryland, College Park, Maryland 20742

and

K. T. Cheng
Argonne National Laboratory, Argonne, Illinois 60439

and

W. R. Johnson
Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556

and

T. B. Lucatorto, T. J. McIlrath,[a] and J. Sugar
Center for Radiation Research, National Bureau of Standards, Washington, D.C. 20234

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The first experimental-theoretical study of Rydberg autoionizing resonances along an isoelectronic sequence is presented. This analysis demonstrates the intimate connection between electron-electron correlation, term dependence, and autoionization and underscores the power of multichannel quantum-defect theory in analyzing complex spectra.

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The combination of the recent development of multichannel quantum-defect theory (MQDT)[1] and the growing capabilities of precision laser spectroscopy has stimulated a renewed interest in the spectroscopy of Rydberg states. These states have provided a rich testing ground in which to refine our understanding of atomic structure,[2] of electric and magnetic field effects on atomic systems,[3] and on radiative interactions.[4] In this paper, we have analyzed the spectra of Xe and Ba^{++} with an empirical MQDT approach and combined this analysis with calculations based on the relativistic random-phase approximation (RRPA)[5] to explain how the changes related to the increasing nuclear charge can be used to reveal some fundamental relationships.

The Rydberg series observed from the $5p^6\,{}^1S_0$ ground state of a Xe-like system can be described in terms of five interacting channels (with $J=1$) which are conveniently labeled by the jj coupling scheme. We denote these channels by ${}^2P_{3/2}ns_{1/2}$, ${}^2P_{3/2}nd_{3/2}$, ${}^2P_{3/2}nd_{5/2}$, ${}^2P_{1/2}ns_{1/2}'$, and ${}^2P_{1/2}nd_{3/2}'$. The first three channels describe the three discrete (or bound) Rydberg series leading up to the $5p^5\,{}^2P_{3/2}$ (or I_1) limit. The fourth and fifth channels describe two series leading up to the second limit, $5p^5\,{}^2P_{1/2}$ (or I_2). The members of the latter two series which lie above I_1 are degenerate with the continuum states of the first three channels; this degeneracy is what gives rise to the autoionization observed between I_1 and I_2.

Figure 1 shows these (Beutler-Fano[7]) autoionizing resonances in a segment of the observed photoabsorption spectra of Xe and Ba^{++} as functions of the effective quantum number ν_2, defined by the relationship

$$E = I_2 - \zeta^2 \mathcal{R}/\nu_2^2, \qquad (1)$$