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Measurement of a Total Atomic-Radiator–Perturber Scattering Cross Section

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From work on the $2S$-$2P_{1/2}$ transition of atomic ^{7}Li, perturbed by noble gases, and use of the photon-echo technique, the first measurement of a "total" atomic-radiator–perturber scattering cross section is reported. The phase-changing, inelastic, *and velocity-changing* aspects of collisions contribute to this cross section, which is significantly larger than the corresponding pressure-broadening cross section. Typical velocity changes are found to be roughly one percent of the mean thermal speed.

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In most spectroscopy experiments, one monitors the dipole moment of the system under investigation. The collisional perturbation of optical dipoles or "optical radiators" represents an interesting problem, since it requires one to understand the way in which collisions affect a superposition state. At first glance, it might seem than any collision destroys the superposition state since the states a and b involved in the optical transition generally follow different collision trajectories.[1,2] The notion of distinct trajectories, however, is a classical one which is known to fail for large-impact-parameter collisions. Thus the dipole moment or optical coherence is not necessarily destroyed in such large impact-parameter collisions.[3] Despite the fact that state-dependent trajectory effects seem to play a crucial role in determining the fate of the optical dipoles, for reasons to be discussed below, steady-state spectroscopy experiments are not overly sensitive[4] to such effects. As a result traditional theories of pressure broadening,[5] in which collisions are assumed to affect only the phases of the optical dipoles, have been successful in explaining these experiments. Only recently has the effect of velocity-changing collisions been put in better perspective.[6] To identify clearly the effects of velocity-changing collisions on optical dipoles experimentally, coherent transient techniques offer unique possibilities.[7]

We present here results of a photon-echo study of $2S$-$2P_{1/2}$ Li radiators perturbed by noble-gas atoms which provide *the first comprehensive picture of the quantum-mechanical velocity-changing aspect of collisions*.[3,7] We measure a total radiator-perturber scattering cross section σ_t (representing the combined effect of the inelastic, phase-changing, and velocity-changing aspects of collisions), and find that it is significantly larger than the broadening cross sections deduced from spectral line measurements. Fitting our data by a phenomenological collision kernel allows us to estimate the average velocity change experienced by a radiator in those collisions which produce identifiable velocity changes.

Assume that the two photon-echo-excitation pulses propagate along $\hat{z}$ and occur at the times $t=0$ and $t=\tau$. The phase of a radiator residing at a particular location $\vec{r}$ in the sample is given[8] (for $t>\tau$) by $\exp[-i(\varphi-\vec{k}\cdot\vec{r})]$, where

$$\varphi(t) \equiv -\int_0^\tau [\omega(t')+\vec{k}\cdot\vec{v}(t')]dt' + \int_\tau^t [\omega(t')+\vec{k}\cdot\vec{v}(t')]dt', \tag{1}$$

$\omega(t')$ $[\vec{v}(t')]$ is the instantaneous oscillation frequency [velocity] of the radiator, and $\vec{k}$ is the common wave vector of the excitation pulses. In the absence of collisions, $\omega(t')$ and $\vec{v}(t')$ are time independent so that $\varphi(t_e=2\tau)=0$ (i.e., Doppler dephasing is eliminated) and an echo is emitted along $\hat{z}$.

In the presence of collisions, the echo intensity I_e is degraded by the factor $\langle \exp[-i\varphi(2\tau)]\rangle^2$, where the angle brackets indicate an ensemble average. While a more detailed calculation of the collisionally induced modification of the echo amplitude will be given elsewhere, we can, roughly speaking, evaluate this factor by considering collisions to be divided into two groups, i.e., those having impact parameters, b, less than or greater than the Weisskopf radius b_W.[5] For $b<b_W$, classical trajectory notions are valid and because of the separation-of-trajectories argument, one is led to the conclusion that the dipole moment is destroyed in a collision. In traditional pressure-broadening theories, phase changes in this region are large enough to destroy the optical coherence. A broadening cross section σ_B can be calculated using either theoretical picture (loss of dipoles because of phase changes or distinct trajectories) and, interestingly, both approaches lead to the same value. For collisions with $b>b_W$, the distinct-trajectory argument fails and one must perform a quantum mechanical calculation. Collisions for which $b>b_W$ give rise to all velocity-changing effects in which the dipole moment may be preserved and the small phase changes which occur in these collisions (which for simplicity we neglect) contribute to the pressure-induced shifts in spectral profiles. We can understand the success of traditional theories of pressure broadening despite their neglect of velocity-changing effects by noting that collisions with $b>b_W$ generally give rise only to small-angle $(\theta \lesssim \lambda_B/b_W \ll 1$, where λ_B is the deBroglie wavelength of Li) diffractive scattering. The small velocity changes associated with diffractive scattering do not significantly modify the output of most steady-state spectroscopic experiments. For large τ, however, photon-echo experiments can be sensitive to the velocity changes resulting from even diffractive scattering.

Thus collisions with $b<b_W$ can be viewed as "inelasticlike" and accordingly lead to a decrease in echo intensity by the factor

$$\exp(-4nv_r\sigma_B\tau), \tag{2}$$

where n is the perturber number density, $v_r = (8k_B T/\pi\mu)^{1/2}$ is the mean perturber-radiator relative speed, k_B is Boltzman's constant, T is the absolute temperature, μ is the radiator-perturber reduced mass, and σ_B is the τ-independent broadening cross section. Velocity-changing collisions decrease the echo intensity by the factor[9]

$$\exp[-4nv_r\tau\sigma_v(\tau)], \tag{3a}$$

where

$$\sigma_v(\tau)=\sigma_v^0\{1-\tau^{-1}\int_0^\tau d\tau' \int_{-\infty}^{\infty} \exp[ik\tau'(\Delta v_z)]g(\Delta v_z)d(\Delta v_z)\}, \tag{3b}$$

σ_v^0 is the total cross section for velocity changing collisions, $k=|\vec{k}|$, and the collision kernel $g(\Delta v_z)$ gives the probability of a particular change in the z component of velocity Δv_z. Over all, the echo intensity varies as

$$I_e(n,\tau)=I_e^0\exp[-4nv_r\tau\sigma_{\mathrm{eff}}(\tau)], \tag{4a}$$

where

$$\sigma_{\mathrm{eff}}(\tau)=\sigma_B+\sigma_v(\tau), \tag{4b}$$

and I_e^0 is the $n=0$ echo intensity. Since the root mean square change in Δv_z is finite for any realistic $g(\Delta v_z)$, $\sigma_v(\tau)=\alpha\tau^2$ for sufficiently small τ

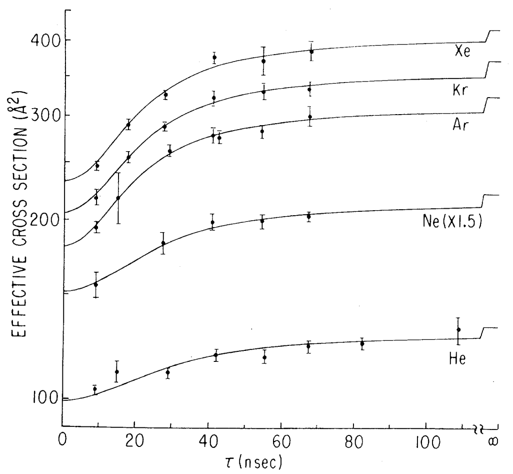

FIG. 1. Plot of $\sigma_{\text{eff}}(\tau)$ vs τ. Error bars represent statistical uncertainty. At right we show σ_t.

and

$$\sigma_{\text{eff}}(\tau) = \sigma_B + \alpha\tau^2 \text{ (short } \tau). \quad (5)$$

Here α depends on the details of $g(\Delta v_z)$. Therefore, $\sigma_{\text{eff}}(\tau = 0) \equiv \sigma_B$. On the other hand, $\sigma_v(\tau \to \infty) = \sigma_v^0$, which implies that $\sigma_{\text{eff}}(\tau \to \infty) = \sigma_t \equiv \sigma_B + \sigma_v^0$.

The excitation pulses used in our experiment on the 6708-Å $2S$-$2P_{1/2}$ transition of ^{7}Li have a 6-GHz spectral width, a 4.5-nsec temporal width, and a peak power of a few watts. The two pulses, optically split from the single output pulse of an N_2-laser–pumped dye laser, were collimated to a 4-mm diameter, and directed through a stainless-steel heat-pipe–type cell containing Li of natural isotopic concentrations. The cell, maintained at 525 ± 15 K (implying a Li pressure of $\approx 10^{-6}$ Torr), had a vapor region approximately 10 cm in length. For reasons described elsewhere,[3] we used excitation pulses of orthogonal linear polarization in short-τ measurements. Excitation pulses of parallel polarization, which, depending on τ, produce echo signals 10 to 100 times larger than pulses of orthogonal polarization, were used for long-τ measurements, where radiative decay of the $2P_{1/2}$ state weakened the echo signal (by ≈ 1600 times for $\tau \approx 100$ nsec). Measurements in the intermediate-τ regime revealed no polarization dependence of observed cross sections.

We have measured I_e vs n (maximum $n \approx 10^{16}$ cm^{-3}) for various fixed τ, and used Eq. (4a) to calculate $\sigma_{\text{eff}}(\tau)$. In Fig. 1, we plot $\sigma_{\text{eff}}(\tau)$ vs τ for each noble-gas perturber. We obtained values of σ_B by using our two shortest-τ measurements of $\sigma_{\text{eff}}(\tau)$ for each perturber and extrapolating, according to Eq. (5), back to $\sigma_{\text{eff}}(\tau = 0) = \sigma_B$. These values of σ_B are presented in Table I along with the values of σ_B obtained in traditional line-broadening experiments.[10] Except in the case of He, we find that the measurements are in good agreement.

We attempt to reproduce $\sigma_v(\tau)$ [computed using our value of σ_B and Eq. (4b)] and hence $\sigma_{\text{eff}}(\tau)$ by inserting various $g(\Delta v_z)$ in Eq. (3b). For $g(\Delta v_z) = (\pi u_0)^{-1/2} \exp(-\Delta v_z^2/u_0^2)$, we obtain the solid lines shown in Fig. 1. The least-squares-fit values of the free parameters σ_v^0 and u_0 are shown in Table I. The quality of the fits is quite good. A Lorentzian kernel of the form $g(\Delta v_z) = (u_0/\pi)/(u_0^2 + \Delta v_z^2)$ [but with $g(\Delta v_z) = 0$ for $\Delta v_z \gg u_0$] produces a better fit to $\sigma_v(\tau)$ for Li-He collisions, but a poorer fit for the other perturbers. These results, as well as the ratio $\sigma_B/\sigma_v^0 \approx 1.0$ observed in our experiment, are in qualitative agreement with a theory based on a quantum mechanical hard-sphere model of collisions.

The derived values of u_0 and σ_v^0 depend somewhat on our choice for $g(\Delta v_z)$. We note, however, that our measurements are sensitive to all velocity changes $\Delta v_z > \Delta v_z^{\min} \equiv 1/k\tau_{\max}$ ($\tau_{\max}$ is our

TABLE I. Various cross sections involved in this work (see text). The G and L in parentheses indicate results obtained with a Gaussian and a Lorentzian collision kernel, respectively.

Perturber	σ_B [a] (Å^2)	σ_B [b,c] (Å^2)	σ_v^0 (G) (Å^2)	σ_v^0 (L) (Å^2)	u_0 (G) (cm/sec)	u_0 (L) (cm/sec)	σ_a [d] (Å^2)	σ_t (G) (Å^2)	σ_t (L) (Å^2)
He	99	86(3)	34	49	1060	247	130	133	148
Ne	101	104(4)	47	90	1140	187	150	149	191
Ar	181	164(8)	145	207	1400	340	400	326	388
Kr	206	211(9)	170	236	1320	335	440	376	442
Xe	233	265(10)	200	289	1320	315	510	433	522

[a]This work.
[b]Scaled from 628 to 525 K according to $(1/T)^{1/5}$.
[c]Ref. 10.
[d]Ref. 11.

maximum pulse separation). Our value for $\Delta v_z^{\min}$ is less than the characteristic diffractive velocity change $v_D \equiv v_r \lambda_B/\sqrt{\sigma_t}$. The fact that the scattering is diffractive in nature restricts our choice of $g(\Delta v_z)$ to functions which are relatively flat in the region $\Delta v_z \lesssim v_D$. With this restriction, we do not expect our derived value of σ_v^0 to differ greatly from those shown in Table I. If a heavier radiator is used, the corresponding values of $u_0 \approx v_D$ could be smaller and the region in which velocity changes are seen (i.e., $ku_0\tau \geqslant 1$) might no longer be accessible. It is perhaps for this reason that velocity-changing effects were not seen in a coherent-transient experiment with I_2 as the radiator.[12]

Recent treatments of radiator-perturber scattering have shown that the net polarization in a medium obeys a quantum mechanical transport equation.[13,14] This equation contains a loss term whose real part, which corresponds to the time rate of change of the polarization's magnitude, is given by $nv_r(\sigma_a+\sigma_b)/2$, where σ_a (σ_b) corresponds to the cross section for ground-state (excited-state) radiator-perturber scattering. Since this is the only term important to the long-time behavior of the photon echo, we equate our σ_t with $(\sigma_a+\sigma_b)/2$.

The cross section σ_a has been measured by atomic-beam techniques.[11] In Table I, we list, for each perturber, the velocity-selected value of σ_a obtained at the velocity v_r. Our values of $\sigma_t=\sigma_B+\sigma_v^0$, based on both Lorentzian and Gaussian kernels, are also presented. Since the long-range part of the potential should, to a reasonable approximation, determine the magnitude of the scattering cross section, we expect (at least for Ar, Kr, and Xe) that $\sigma_b/\sigma_a \approx (C_6^b/C_6^a)^{2/5}$, where C_6^b (C_6^a) is the coefficient of the Van der Waals portion of the potential. Assuming that $C_6 = 2\epsilon r_m^6$, where ϵ is the potential well depth and r_m is the internuclear separation at which the potential is minimum [as is true of a Lennard-Jones (6,12) potential], we can use the potential calculation of Baylis[15] to find that $(C_6^b/C_6^a)^{2/5}$ falls between 1 and 1.6, depending on the perturber and which of the possible excited-state potentials is considered. This implies that σ_t should be between 1 and 1.3 times σ_a. Our data are reasonably consistent with this result.

Our results clearly demonstrate the role played by velocity-changing collisions. Their contribution to the echo signal underlines the importance of quantum mechanical scattering effects in collisions undergone by an optical dipole.

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