or holes broken ($\Delta\delta = 0$ for $J^\pi = \frac{1}{2}^-$), so that our data should lie on a straight line. (The results deduced from our tables are consistent with these predictions.) Finally, let us note that these pseudo mirror pairs all satisfy the relations

$$[\mu(t, t_3) - J]/\mu(t, -t_3) = -1.20,$$

within a few percent. This is an enlargement of the so-called De Shalit relation $(g_s - g_l)_p/(g_s - g_l)_n = -1.20$ deduced in the frame of the extreme single-particle model.

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Total Scattering Cross Section for Na on He Measured by Stimulated Photon Echoes

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A technique which employs heretofore unappreciated aspects of the optical stimulated echo was developed and used to measure the total scattering cross section for Na($3S_{1/2}$)-He and Na($3P_{1/2}$)-He collisions. The repulsive-core–dominated Na($3S_{1/2}$)-He collision is found to be relatively soft. The cross section for degradation of the $3S_{1/2}$-$3P_{1/2}$ superposition, determined from photon-echo–decay measurements, is anomalous in that it is smaller than the total elastic-scattering cross section with Na in either state separately.

The study of atomic scattering, traditionally carried out by atomic-beam measurements,[1] has recently been supplemented by laser techniques.[2] In this Letter we report the first application of a coherent-transient laser technique to the study of velocity-changing collisions (VCC) in an atomic system.[3] Our technique utilizes unappreciated novel aspects of the three-pulse stimulated photon echo[4] (SP echo) to provide relatively direct measurements of VCC. We predict and demonstrate experimentally (e.g., Fig. 1) that phase memory information stored in *either* the ground

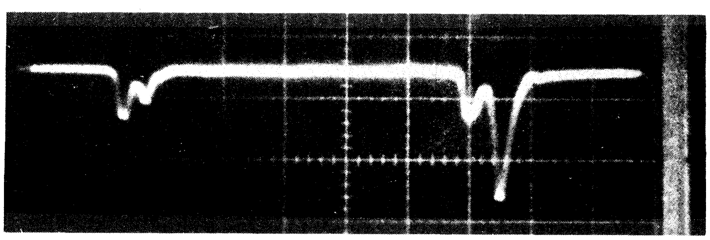

FIG. 1. Oscilloscope trace showing scattered light from three excitation pulses (third row of the table in Fig. 3 with $\vec{K}'\cdot\hat{z}/|\vec{K}'| = -1$) and the subsequent SP echo (fourth pulse) pronounced on the $3S$-$3P_{1/2}$ transition of Na when the second-to-third pulse separation is $\cong 17$ times the 16-nsec lifetime of the $3P_{1/2}$ state. At the time of the third pulse a negligible $\cong 10^{-8}$ of the intially excited population remains in the $3P_{1/2}$ state. The echo persists because of the information stored in the ground state alone. (Horizontal: 50 nsec/div.)

or the excited state of the echo transition can be separately recalled by the third excitation pulse to generate an SP echo. This marks the first report of an echo generated from information stored in a *single state*. In the second-to-third–pulse interval, t_{32}, the SP "echo information" is *not* stored in a superposition state. Thus the phase-changing aspect of collisions, which tends to make VCC measurements difficult with other laser techniques, is completely irrelevant during t_{32}. The SP echo, generated from information stored in a single state (Fig. 1), isolates VCC affecting that state *alone*. The SP-echo technique, which among other things provides the absolute total-scattering cross section, should be particularly useful for measurements involving nonmetastable excited states where atomic-beam techniques are difficult to apply.

The SP echo is treated in general elsewhere.[5] We concentrate here on essential new features. Consider a Doppler-broadened gaseous sample of "echo" atoms. Each atom has a nondegenerate ground state ($|\alpha\rangle$) and excited state ($|\beta\rangle$); only $|\alpha\rangle$ is initially populated. The three, short, SP-echo excitation pulses occur at times t_i, and have wave vectors $\vec{k}_i$ ($i=1,2,3$), and pulse areas of $\pi/2$. The first two pulses with $\vec{k}_1=\vec{k}_2\equiv\vec{K}\parallel\hat{z}$ couple $|\alpha\rangle$ and $|\beta\rangle$ and generate diagonal elements of ρ (the resulting density matrix) given by

$$\rho_{\alpha\alpha}=\sin^2(Kv_z t_{21}/2),\quad \rho_{\beta\beta}=\cos^2(Kv_z t_{21}/2),$$

where v_z is the z component of the thermal velocity and $t_{ij}=t_i-t_j$.[5] Since $\rho_{\alpha\alpha}$ and $\rho_{\beta\beta}$ contain no information regarding the sense of $\vec{K}$ along $\hat{z}$, and since, as is well known, the SP-echo information resides *solely* in the diagonal elements of ρ during t_{32}, it is clear that the echo, which has wave vector $\vec{k}_e=\vec{k}_3\equiv\vec{K}'$, will be produced whether $\vec{k}_3$ is parallel or antiparallel to $\vec{K}$. This important feature has not been previously recognized.

Since only the diagonal elements of ρ are relevant after t_2, it follows that the action of the third pulse can be understood in terms of a free decay.[5] We generalize previous results by assuming that pulse 3 couples levels $|\beta\rangle$ and $|\gamma\rangle$, where $|\gamma\rangle$ is another initially unpopulated excited state. The free decay field emitted on the $|\beta\rangle$-$|\gamma\rangle$ transition is calculated in the standard way from

$$E_{\text{tot}}=\int_{-\infty}^{\infty}n(v_z)\rho_{\beta\beta}E(v_z,t)\,dv_z,$$

where $n(v_z)$ is the thermal v_z distribution, $E(v_z,t)\propto\exp[-i(\omega_{\beta\gamma}\pm k_3v_z)t]$ is the Doppler-shifted field emitted from atoms of velocity v_z, and $\omega_{\beta\gamma}$ is the unshifted $|\beta\rangle$-$|\gamma\rangle$ transition frequency.[5] With use of the $\rho_{\beta\beta}$ given above this yields an echo on the $|\beta\rangle$-$|\gamma\rangle$ transition at the time $t_e=Kt_{21}/k_3+t_3$.[6] This constitutes the first demonstration of an echo created by information stored in a single atomic state.

In the simple model above the relaxation of atoms in $|\beta\rangle$ back to $|\alpha\rangle$ returns the sample to its preexcited thermal equilibrium state since $\rho_{\alpha\alpha}+\rho_{\beta\beta}=1$. In the presence of magnetic degeneracy and/or hyperfine structure, however, alternative decay channels prevent the sample from returning to thermal equilibrium immediately; consequently, even after all atoms in $|\beta\rangle$ have radiatively decayed, a third pulse coupling $|\alpha\rangle$ and $|\beta\rangle$ can still produce an echo from the information remaining in $|\alpha\rangle$. An echo generated in such a fashion was used to obtain the data of Fig. 2. Magnetic degeneracy also has the effect of allowing the echo and excitation pulses to have different polarizations. (The relative polarizations observed here are shown in Fig. 3.)

With the assumption that the SP echo is generated from the information stored in one state only, VCC between echo atoms and perturber atoms during t_{32} degrade the echo intensity, I_e, accord-

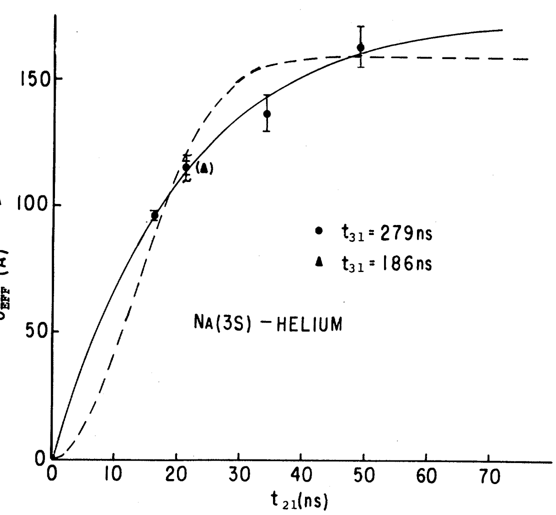

FIG. 2. The effective scattering cross section, σ_{eff}, for Na(3S)-He collisions plotted vs $t_{21}=t_{e3}$. The error bars represent the statistical uncertainty of our data. The solid curve was generated using a Lorentzian scattering kernel (see text) with $\Delta v_0=460$ cm/sec. For the Lorentzian curve $\sigma_{\text{eff}}(t_{21}\to\infty)=\sigma_{1\text{im}}=176$ Å^2. The dashed line was generated using a Gaussian kernel with $\Delta v_0=1050$ cm/sec and $\sigma_{\text{lim}}=159$ Å^2. The 21-nsec points indicate that within experimental error σ_{eff} is independent of t_{31}.

PULSE POLARIZATIONS 1	2	3	ECHO	$\vec{K}\cdot\hat{z}/\lvert\vec{K}\rvert$	$\vec{K}'\cdot\hat{z}/\lvert\vec{K}'\rvert$
↑	↑	↑	↑	1	±1
↑	→	↑	→	1	±1
↑	→	→	↑	1	±1

FIG. 3. SP echoes have been observed on the $3S$-$3P_{1/2}$ transition of Na produced by excitation pulses having all combinations of linear polarizations and propagation directions shown here. The echo was observed through a polarizer oriented as shown.

ing to $\langle\exp(-iK'\Delta v_T t_{e3})\rangle^2$, where Δv_T is the *total* change in v_z experienced by a given atom during t_{32} (as the result of many VCC) and the brackets denote an average over all atoms at a particular point in the sample when $t=t_e$. If all collisions are uncorrelated, and the probability that a single VCC will result in a change in v_z by Δv is independent of the initial velocity, then the contribution to this average from l collisions is

$$\exp(-\Gamma t_{32})[\Gamma t_{32}\langle\langle\exp(-iK'\Delta v t_{e3})\rangle\rangle]^l/l!,$$

where $\Gamma=nv_r\sigma$ is the rate at which collisions occur, n is the number density of perturber atoms, $v_r=(8k_B T/\pi\mu)^{1/2}$, k_B is the Boltzmann constant, T is the absolute temperature, μ is the reduced mass of the echo and perturbing atom, σ is a thermally averaged total VCC (elastic scattering) cross section, and the double angular brackets denote an average over Δv weighted by the probability that a change in v_z of Δv will occur in a single VCC.

Summing this expression over l we have $\langle\exp(-iK'\Delta v_T t_{e3})\rangle^2=\exp(-\beta' P)$, where P is the perturber-gas pressure, $\beta'=2(n_0/P_0)v_r\sigma_{\rm eff}t_{32}$, where

$$\sigma_{\rm eff}=\sigma[1-\langle\langle\exp(-iK'\Delta v t_{e3})\rangle\rangle],$$

and n_0 is the perturber-gas density at pressure P_0.[7] For t_{e3} so large that, $K't_{e3}\langle\langle|\Delta v|\rangle\rangle\gg 1$, β' approaches $2(n_0/P_0)v_r\sigma t_{32}$, $\sigma_{\rm eff}\to\sigma$, and the total cross section can be unambiguously determined. The behavior of $\sigma_{\rm eff}$ in the short-time regime, where $\sigma_{\rm eff}<\sigma$, can be calculated by assuming a scattering kernel $f(\Delta v)$ such that

$$\langle\langle\exp(-iK'\Delta v t_{e3})\rangle\rangle = \int_{-\infty}^{\infty} d(\Delta v) f(\Delta v)\exp(-iK'\Delta v t_{e3}).$$

Intuitively, the dependence of $\sigma_{\rm eff}$ (the effective echo-decay cross section for VCC during t_{32}) on the interval t_{e3} or t_{21} can be seen as follows: The VCC have a characteristic $\langle\langle|\Delta v|\rangle\rangle$. The "modulation" period of $\rho_{\alpha\alpha}$ and $\rho_{\beta\beta}$ versus v_z depends on $t_{21}\propto t_{e3}$. Finally, the effectiveness of the average Δv in destroying (thermalizing) the modulation depends on the ratio $\langle\langle|\Delta v|\rangle\rangle$ ("modulation" period).

Over all the echo intensity I_e decays with pressure as $\exp(-\beta P)$, where $\beta=\beta'+\beta''$,

$$\beta''=2(n_0/P_0)v_r(\sigma_{21}t_{21}+\sigma_{e3}t_{e3}),$$

and σ_{ij} is the cross section for collisional decay of the atomic superposition state relevant to the echo during the interval t_{ij}.

Our experiments have been performed primarily on the $3S_{1/2}$-$3P_{1/2}$ transition of Na vapor. The Na is contained in a heat-pipe-type cell at 400 ± 15 K. Since the vapor pressure of Na is $\cong 10^{-5}$ Torr, Na-Na collisions are insignificant. The foreign-gas (He) pressure P (always <0.5 Torr) is measured by a capacitance manometer. The excitation pulses are supplied by one (or two) N_2-laser–pumped dye laser(s) of ≅ 1 GHz spectral and 7 nsec temporal width. The Glan-prism–(GP-) polarized, collimated excitation pulses have an ≅ 2 mm diam and peak power of a few watts in the Na cell. A dual optical-delay line is used to vary t_{21} and t_{31} independently. *SP echoes were observed with all the pulse polarizations and relative propagation directions shown in Fig. 3.* These measurements were all made with $t_{32}>10\tau$, where $\tau=16$ nsec is the $3P_{1/2}$ state lifetime. When excitation pulses polarized parallel to the echo copropagate with the echo, it is necessary to protect the echo-detecting photomultiplier tube (PMT) with a series of electro-optic shutters (EOS) gated to pass at the time of the echo. On the other hand, when $\vec{K}'\cdot\hat{z}/|\vec{K}'|=-1$ and pulse 3 is polarized orthogonal to the echo, *a GP polarizer crossed with respect to pulse 3 sufficiently reduces the background level ($\times 10^6$) to allow the echo to be seen.* To improve our discrimination we also inserted a single EOS before the PMT. Unless otherwise specified our measurements were made using SP echoes produced by the excitation sequence shown in the bottom row of the table in Fig. 3 with $\vec{K}'\cdot\hat{z}/|\vec{K}'|=-1$.

Our cross-section measurements are obtained from the $\exp(-\beta P)$ decay of I_e for various t_{21} and t_{31}. For our $3S$-$3P_{1/2}$ transition measurements, where $K=K'$, the factor $\exp(-\beta'' P)$ is the pressure-dependent decay of a two-pulse photon echo[8] (PE) produced on the $|\alpha\rangle$-$|\beta\rangle$ transition by two pulses separated by t_{21}. We therefore use our PE decay data[9] to obtain β'' and determine $\beta'=\beta-\beta''$.

σ_{eff} is then obtained from

$$\sigma_{eff} = (1.04\times 10^{-19}\ \mathrm{cm}^3\ \mathrm{Torr/K})\beta' T/2t_{32}v_r.$$

Since population is only present in the $3^2S_{1/2}$ state at the time of the third pulse, σ_{eff} represents the cross section for Na(3S)-He collisions.

The values of σ_{eff} derived from our measurements of Na(3S)-He collisions are shown in Fig. 2 as a function of t_{21} (t_{31} fixed). As expected, σ_{eff} increases rapidly for small t_{21} and then flattens out. At $t_{21} = 49$ nsec we obtain $\sigma_{eff} = 163 \pm 9$ Å^2 as a lower limit for σ. Our value lies between the values of $\sigma = 130$ and 247 Å^2 measured elsewhere.[10] All data points except the triangular one were taken with $t_{31} = 279$ nsec. The triangular point, obtained for $t_{31} = 186$ nsec, falls within the error bars of the 279-nsec point. The solid curves shown in Fig. 2 were calculated for

$$f(\Delta v) = (\Delta v_0/\pi)(\Delta v_0{}^2 + \Delta v^2)^{-1};$$

the dashed, for

$$f(\Delta v) = (\Delta v_0 \pi^{1/2})^{-1} \exp(-\Delta v^2/\Delta v_0{}^2).$$

The region of the interatomic potential to which a given cross-section measurement is most sensitive is determined[11] by the ratio v_c/v_r ($v_c = 2\epsilon r_m/\hbar$, where ϵ is the depth of the attractive potential well, and r_m is the interatomic distance at which the potential is minimum). For Na(3S)-He ($\epsilon \cong 2$ cm^{-1}, $r_m \cong 5.6$ Å),[12] v_c/v_r is only 0.3, which implies that the repulsive core dominates the scattering interaction. Measurements of Li(2S)-He scattering by Dehmer and Wharton[13] indicate that the repulsive core is quite "hard." Thus, since the Li(2S)-He and Na(3S)-He potentials are expected to be similar,[12] it is tempting to describe the Na(3S)-He collisions by a hard-sphere model. Such a model has been developed by Borenstein and Lamb,[14] who find that for hard-sphere collisions the average velocity change, Δv_0, is of order v_r and that the collision kernel is of Gaussian form. However, we find that for any reasonable $f(\Delta v)$ our data force us to conclude that $\Delta v_0 \ll v_r$ ($= 1.6\times 10^5$ cm/sec). Furthermore, Fig. 2 shows that a Gaussian kernel cannot properly simulate our data. Thus, although the Na(3S)-He potential is believed to be relatively "hard," we see that the entire spectrum of Na(3S)-He collisions cannot be characterized as "hard-sphere."

It is interesting to note that the Lorentzian kernel describes our predominantly small-Δv data quite well. The limiting cross section obtained with the Lorentzian kernel with use of $\Delta v_0 = 460$ cm/sec is only 8% greater than the lower limit mentioned above.

Using SP echoes (generated as shown in the first row of the table in Fig. 3 with $\vec{K}'\cdot\hat{z}/|\vec{K}'| = 1$), we have made preliminary measurements of σ for Na(3$P_{1/2}$)-He collisions. In this case the first two excitation pulses ($t_{21} = K't_{e3}/K = 33$ nsec) are resonant with the $3S_{1/2}$-$3P_{1/2}$ transition, while the third ($t_{31} = 56$ nsec) is resonant with and creates an echo on the $3P_{1/2}$-$4D_{3/2}$ transition. Using PE[9] and excited-state PE[15] measurements to obtain β', we find that

$$\sigma(\mathrm{Na}(3P_{1/2})\text{-He}) \cong 2.5\sigma(\mathrm{Na}(3S)\text{-He}).$$

We note that PE measurements[9] yield the cross section $\sigma_{21} = 129 \pm 4$ Å^2 for He-induced decay of the $3S$-$3P_{1/2}$ superposition.[16] This result is anomalous, since it is smaller than the VCC cross section associated with either the 3S or 3$P_{1/2}$ states separately. The implications of this result are currently being explored.

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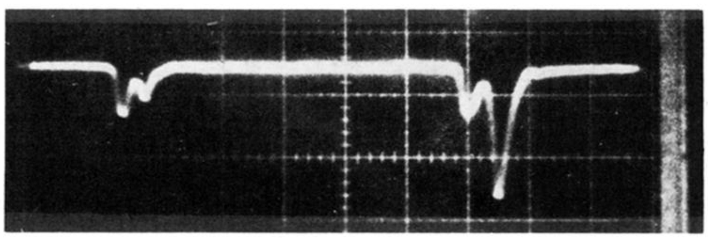

FIG. 1. Oscilloscope trace showing scattered light from three excitation pulses (third row of the table in Fig. 3 with $\vec{K}'\cdot\hat{z}/|\vec{K}'| = -1$) and the subsequent SP echo (fourth pulse) pronounced on the $3S$-$3P_{1/2}$ transition of Na when the second-to-third pulse separation is $\cong 17$ times the 16-nsec lifetime of the $3P_{1/2}$ state. At the time of the third pulse a negligible $\cong 10^{-8}$ of the intially excited population remains in the $3P_{1/2}$ state. The echo persists because of the information stored in the ground state alone. (Horizontal: 50 nsec/div.)