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Tri-Level Echoes

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We report a new Doppler-free rephasing effect, the *tri-level echo,* which we use to study Ar-Na collisional relaxation of several $3^2S_{1/2}$-$n^2D_{3/2}$ superpositions in atomic Na. Three excitation pulses are required: The first resonantly excites a selected state, while the others resonantly couple this state with a higher-lying state. This sequence produces a delayed rephasing on the resonance transition which radiates strongly; the radiated intensity monitors relaxation in the higher-lying stepwise-excited state.

It is well known that an echo, i.e., a rephasing of superposition states, may be formed in a sample of inhomogeneously broadened two-level systems. First observed by Hahn in spin systems (spin echoes)[1] and later by Abella, Kurnit, and Hartmann in systems of electronic states (photon echoes),[2] these two-level single-frequency echoes have proven useful in the study of homogeneous relaxation. This basic phenomenon has recently been extended by the observation[3] of the Raman echo,[4] which is similar to the photon echo except that the two states are connected by a two-photon interaction matrix element. In this Letter we present a new type of echo peculiar to multilevel (three levels or more) systems. We have discovered that for a sample of multilevel systems inhomogeneously broadened by the Doppler effect[5] a rephasing of a coherent superposition between a particular pair of levels can be induced even if the constituent systems of the sample spend most of the time between their first excitation and the echo dephasing in a superposition between a *different* pair of levels. The echo signals produced are large since they arise on an allowed transition. Through them one can study the relaxation of a step-wise, resonantly excited superposition which would ordinarily be inaccessible. We term this effect *tri-level echoes*. Similar effects were predicted several years ago by Aihara and Inaba,[6] who described them as anomalous photon echoes. Related effects were observed in three-level spin systems by Hatanaka, Terao, and Hashi.[7] We have observed tri-level echoes in Na on the $3^2S_{1/2}$-$3^2P_{1/2}$-$n^2D_{3/2}$ (n =4, 6, 7, 8, and 9) three-level systems, and we have used it to measure the Ar-Na collisional decay constant of the Na $3^2S_{1/2}$-$4^2D_{3/2}$ and $3^2S_{1/2}$-$7^2D_{3/2}$ superpositions.

Consider a gaseous sample of three-level systems whose atomic states $|0\rangle$, $|1\rangle$, and $|2\rangle$ have respective eigenenergies $\hbar\Omega_0$, $\hbar\Omega_1$, and $\hbar\Omega_2$ ordered for simplicity as $\Omega_0<\Omega_1<\Omega_2$. The atoms are initially in state $|0\rangle$. The transitions 0-1 and 1-2 are $E1$ allowed. The sample is irradiated at times $t_1 \leq t_2 < t_3$; the ith excitation pulse has central frequency ω_i and wave vector $\vec{k}_i$. We specialize to $\omega_1=\Omega_{10}$ and $\omega_2=\omega_3=\Omega_{21}$, where $\Omega_{ij}=|\Omega_i-\Omega_j|$.[8] Under these conditions, complete rephasing on the resonance transition producing an echo at $\omega_4=\omega_1+\omega_2-\omega_3$ will occur at $t=t_4$ when and if all atoms at $\vec{x}$ are in superposition states of identical relative phase φ and are phase matched according to $\varphi(\vec{x})=\vec{k}_4\cdot\vec{x}$. In the absence of velocity-changing collisions, an atom having velocity $\vec{v}$ and position $\vec{x}$ at time t must have come from $\vec{x}_i=\vec{x}-\vec{v}(t-t_i)$ at $t=t_i$. The atomic superposition created between states $|0\rangle$ and $|2\rangle$ by excitation pulses 1 and 2 has the phase factor $\exp[i(\vec{k}_1\cdot\vec{x}_1+\vec{k}_2\cdot\vec{x}_2)]$. The third pulse transforms this $|0\rangle$-$|2\rangle$ superposition into a $|0\rangle$-$|1\rangle$ superposition, introducing an additional phase factor $\exp(-i\vec{k}_3\cdot\vec{x}_3)$. For this atom the net phase of the $|0\rangle$-$|1\rangle$ superposition is thus

$$\varphi=\vec{k}_1\cdot\vec{x}_1+\vec{k}_2\cdot\vec{x}_2-\vec{k}_3\cdot\vec{x}_3=\varphi_0-\vec{v}\circ\vec{A}(t), \qquad (1)$$

where

$$\varphi_0=(\vec{k}_1+\vec{k}_2-\vec{k}_3)\circ\vec{x}$$

and

$$\vec{A}(t) = (t - t_1)\vec{k}_1 + (t - t_2)\vec{k}_2 - (t - t_3)\vec{k}_3.$$

Under the assumption that the entire Doppler distribution is equally excited, the dipole moment of frequency ω_4, produced at $\vec{x}$ when $t > t_3$, is proportional to $\langle e^{i\varphi}\rangle$ where the brackets indicate an average over the velocity distribution. Assuming perfect phase matching, i.e., $\vec{k}_4 = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$, the radiated intensity I at ω_4 is proportional to $\exp(-\frac{1}{2}v_0^2|\vec{A}|^2)$, where $v_0 = (2kT/m)^{1/2}$, k is Boltzmann's constant, T is the temperature, and m is the mass of the atom. The intensity peaks when the various atomic dipoles rephase so as to minimize $|\vec{A}(t)|$. Setting $t_1 = 0$, this occurs at the "echo" time t_4 given by $t_4 = (t_2\vec{k}_2 - t_3\vec{k}_3)\cdot\vec{k}_4/|\vec{k}_4|^2$. In terms of t_4, $\vec{A}(t)$ can be expressed as $\vec{A}(t) = (t - t_4)\vec{k}_4 - t_2\vec{k}_{2\perp} + t_3\vec{k}_{3\perp}$, where $\vec{k}_{i\perp}$ is the component of $\vec{k}_i$ normal to $\vec{k}_4$. From the above it should be clear that complete rephasing, i.e., $\vec{A}(t_4) = 0$, can only occur if the $\vec{k}$ vectors are coplanar. A case of special interest is $\hat{k}_1\cdot\hat{k}_2 = -1$ and $\vec{k}_2 = \vec{k}_3$, for which $\vec{k}_4 = \vec{k}_1$ and $\vec{A}(t) = (t - t_4)\vec{k}_4$. Under the assumption that dispersion may be neglected, rephasing will in this case occur at $t_4 = (t_3 - t_2)\omega_2/\omega_1$ as long as $t_4 > t_3$; the latter condition is insured if $\omega_2 > \omega_1 t_3/(t_3 - t_2)$. Since $\vec{k}_4 = \vec{k}_1$, the echo is in this case emitted along the direction of propagation of pulse 1. Noncollinear excitation, however, is desirable in order to avoid saturation of the detection system by the first excitation pulse. Unlike photon echoes, tri-level echoes may easily be phase matched in the noncollinear configuration. Like photon echoes,[9] however, tri-level echoes do not, in general, rephase perfectly when the beams are not collinear. For tri-level echoes perfect rephasing in the noncollinear case, which requires that the quantity $t_2\vec{k}_{2\perp} - t_3\vec{k}_{3\perp}$ vanish, only occurs for certain values of t_2 and t_3 when $\vec{k}_{2\perp}\|\vec{k}_{3\perp}$. For small departures from our case of special interest this quantity is, however, always rather small. We may evaluate the echo intensity I for the geometry of Fig. 1 ($\theta \ll 1$) and find

$$I = I_0 \exp\left(-\frac{\theta^2 v_0^2 \omega_2^2}{2c^2}\left[\frac{(\omega_2 - \omega_1)}{2\omega_1}(t_3 - t_2) - t_2\right]^2\right),$$

where I_0 is the intensity when $\theta = 0$, and where we have neglected the effect of the noncollinearity on the size of the beam-overlap volume. For the relatively unfavorable case of $\omega_2/\omega_1 = 1.5$, with $t_2 = 0$, $I(\theta = 0.02\text{ rad})$ is smaller than $I(\theta = 0)$ by only a factor of 10 at $t_3 = 50$ nsec. Also, for the experimental situations that we have examined, t_4 differs insignificantly from the value it has when the excitation is collinear.

It should be noted that in contrast to a two-pulse, two-level photon echo, a tri-level echo cannot generally be explained simply in terms of a reversal of the relative phase of each atomic superposition between a particular pair of levels.[10] However, using appropriately generalized concepts, an analogous formulation may be useful. We leave elaboration on this topic to a future publication.

In our experiment the Na $3^2S_{1/2}$, $3^2P_{1/2}$, and $n^2D_{3/2}$ states ($n = 4, 6, 7, 8, 9$) correspond to $|0\rangle$, $|1\rangle$, and $|2\rangle$, respectively. The $5^2S_{1/2}$ state has also served as state $|2\rangle$. These states are excited by 7-nsec (full width at half-maximum) FWHM excitation pulses produced by two nitrogen-laser-pumped dye lasers. The first dye laser, of frequency ω_1 resonant with the D_1-line transition, has a 750-MHz spectral width, while the second, of frequency ω_2 ($= \omega_3$), has a 10-GHz spectral width. The output of the second dye laser is split; an undelayed part is used as pulse 2, and the other part is optically delayed as pulse 3. Laser peak power is ~50 W for 3-mm beam diameters. By suitable optics the laser pulses are made to cross inside a sodium cell as shown in Fig. 1. The angle θ of Fig. 1 was usually chosen to lie between 20 and 50 mrad. The cell consists of a 65-cm-long stainless-steel tube with

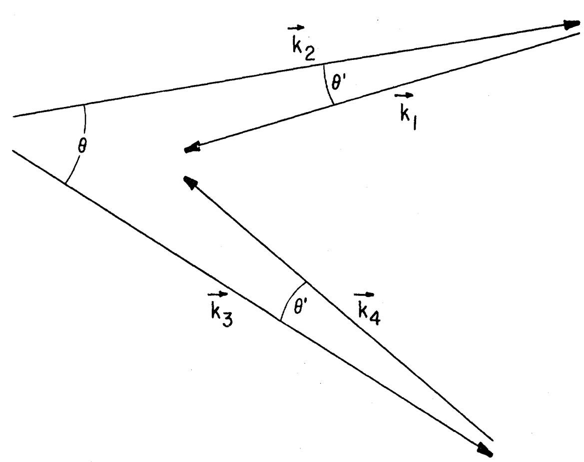

FIG. 1. Relative orientations of the coplanar excitation pulses with propagation vectors $\vec{k}_1$, $\vec{k}_2$, $\vec{k}_3$, and the resulting echo with propagation vector $\vec{k}_4$ in the experimental arrangement used to observe the $3^2S_{1/2}$-$3^2P_{1/2}$-$|2\rangle$ tri-level echoes, where $|2\rangle$ is either the $4^2D_{3/2}$ or the $7^2D_{3/2}$ state. In the case where $|2\rangle$ is the $4^2D_{3/2}$ state, $\theta' < 1$ mrad when $\theta \cong 50$ mrad.

windows on its ends. Foreign gas can be introduced into the cell, and its pressure is measured by a Baratron capacitance manometer. The sodium pressure is determined by its vapor pressure at the oven temperature. The echo, whose frequency is ω_1, propagates nearly antiparallel to $\vec{k}_3$ and is detected on a photomultiplier. The $3^2S_{1/2}$-$3^2P_{1/2}$-$4^2D_{3/2}$ echo typically contains about 10^6–10^7 photons. The echo size decreases quite slowly as we go to higher D states.

When ω_2 is resonant with the 568.3-nm $3^2P_{1/2}$-$4^2D_{3/2}$ transition, $\omega_2/\omega_1 = 1.038$. Thus when $t_1 = t_2 = 0$ and $t_3 = 195$ nsec, $\Delta t = t_4 - t_3$ should be about 7 nsec. We have indeed observed this delay of the echo from the time of the third pulse. A longer delay Δt is expected when ω_2 is resonant with the 449.4-nm $3^2P_{1/2}$-$7^2D_{3/2}$ transition. Here, with $t_1 = t_2 = 0$ and $t_3 = 81$ nsec, Δt should be ~25 nsec. The observed delay of 23 nsec agrees quite well with this value when one considers the 7-nsec pulse widths and the difficulty of making t_1 and t_2 equal. We have also observed that the echo intensity changes when a magnetic field is applied perpendicular to $\vec{k}_4$; this suggests the presence of quantum-beat effects.

In the present experiments the superposition ρ_{10} which produces the echo decays according to $\rho_{10} \sim \exp\{-[\Gamma_{10}(t_2 - t_1) + \Gamma_{20}(t_3 - t_2) + \Gamma_{10}(t_4 - t_3)]\}$. Here Γ_{ij} denotes the total homogeneous decay rate, $1/T_2$, for the $|i\rangle$-$|j\rangle$ superposition. We rewrite Γ_{ij} as $\Gamma_{ij} = \Gamma_{ij}^0 + \eta_{ij}p$, where p is the foreign-gas pressure, $\eta_{ij}p$ is the decay rate due to Na–foreign-gas collisions, and Γ_{ij}^0 is the decay rate of the $|i\rangle$-$|j\rangle$ superposition in the absence of the foreign gas. For our experiments $t_2 \cong t_1 = 0$. The dependence of the echo intensity I on the foreign-gas pressure is therefore $I(p) \sim |\rho_{10}|^2 \sim e^{-\beta p}$, where

$$\beta = 2\{\eta_{20} + \eta_{10}(\omega_2 - \omega_1)/\omega_1\} t_3.$$

With Ar as the foreign gas, we have measured β by varying p at fixed values of t_3. The decay rate η_{20} may thus be determined from a knowledge of β and η_{10}, and the Ar-induced 0-2 transition linewidth $\Delta\nu$(FWHM) is given in hertz/Torr by η_{20}/π. Using our preliminary experimental values of β and the value of η_{10} measured by the photon-echo technique,[11] $(4\eta_{10})^{-1} = 4.2 \pm 0.3$ nsec Torr, we obtain the following Ar-induced linewidths at a temperature $T \cong 410$°K: For the $3^2S_{1/2}$-$4^2D_{3/2}$ transition, $\Delta\nu = 60 \pm 10$ MHz/Torr; and for the $3^2S_{1/2}$-$7^2D_{3/2}$ transition, $\Delta\nu = 160 \pm 20$ MHz/Torr. In the former case no change in $\Delta\nu$ is observed as t_3 is varied from 27 to 106 nsec. The 3S-4D Ar-induced linewidth has been measured by two other recent experiments. Liao, Economou, and Freeman,[12] using a Doppler-free two-photon transient technique, find $\Delta\nu = 47$ MHz/Torr at 670°K, and Biraben *et al.*,[13] using Doppler-free two-photon absorption, find $\Delta\nu = 52 \pm 5$ MHz/Torr at 560°K. For an ideal gas, the value of $\Delta\nu_1$ at temperature T_1 is related to $\Delta\nu_2$ at temperature T_2 by $\Delta\nu_1 = \Delta\nu_2(T_2/T_1)^{1/2}\sigma_1/\sigma_2$, where σ_i the the Ar-Na collisional cross section at temperature T_i; hence the two experiments give 60 MHz/Torr and 61 ± 6 MHz/Torr, respectively, at our temperature if we assume $\sigma_1 = \sigma_2$. Thus, considering the experimental uncertainties, the three values are in good agreement for any reasonable velocity dependence that σ is assumed to have. It is interesting to note that to within experimental uncertainty our values of $\Delta\nu$ are identical with the $3^2P_{1/2}$-$4^2D_{3/2}$ and $3^2P_{1/2}$-$7^2D_{3/2}$ Ar-induced decay measurements made by the excited-state photon-echo technique.[11] This result suggests that the P state plays a small role in the Ar-induced decay of these P-D superpositions.

Other examples of tri-level echoes are the following: Let $\omega_1 = \omega_3 = \Omega_{10}$ and $\omega_2 = \Omega_{21}$. If $\vec{k}_3 = \vec{k}_1$ and either (i) $\vec{k}_1$ and $\vec{k}_2$ are antiparallel, and $\Omega_0 < \Omega_1 < \Omega_2$, or (ii) $\vec{k}_1$ and $\vec{k}_2$ are parallel, and $\Omega_0 < \Omega_2 < \Omega_1$, then an echo of wave vector $\vec{k}_2$ is formed at $t_4 = t_3\omega_1/\omega_2 + t_2$ if $t_1 = 0$ and $t_4 \geq t_3$. Furthermore, if $\Omega_0 < \Omega_1 < \Omega_2$, $\omega_1 = \omega_2 = \Omega_{10}$, $\omega_3 = \Omega_{21}$, $\vec{k}_{1,2,3}$ are all parallel and $t_1 = 0$, then an echo of frequency $\omega_4 = \Omega_{21}$ occurs at $t_4 = t_3 + t_2\omega_2/\omega_3$. We have already observed the latter echo. Many other variations of the basic tri-level echo scheme should be possible. A variety of echoes involving more than three levels ("multilevel" echoes) and/or arising from a sequence of more than three excitation pulses are possible as well. We note that it is also correct to visualize the tri-level echo as delayed Doppler-free four-wave mixing.

In summary, we have observed a new rephasing phenomenon, the tri-level echo. The rephasing, which occurs between two levels connected by an electric-dipole moment in a three-level system, depends for part of the interval between excitation and echo on superpositions which cannot radiate. Thus the properties of these nonradiating superpositions can be studied. Unlike Raman echoes or the sum-frequency analog of Raman echoes, which provide information about the same type of superpositions, tri-level echoes are produced primarily by on-resonance excitation. This means that low laser powers are required. Unlike excited-state photon echoes which tend to become weaker as the oscillator strength of the excited transition decreases,[11] tri-level echoes should remain of constant intensity as long as the lasers can produce 180° pulses for the transitions involved. Reference 11 shows how modest these power requirements are, and it is thus reasonable to believe that tri-level echoes will be observable when state $|2\rangle$ is a highly excited state in virtually any element. Tri-level echoes should thus be an extremely powerful tool for relaxation studies in both pulsed and cw experiments. Finally, we mention one surprising observation: Under the conditions described in the paragraph preceding Eq. (1), with $t_1 = t_2 = 0$, an undelayed signal at $\omega = \omega_1$ occurs at $t = t_3$ when state $|2\rangle$ is the $5^2S_{1/2}$ state and $\hat{k}_1 \cdot \hat{k}_2 = -1$. In this case no echo should appear according to the analysis following Eq. (1), since $\omega_2 < \omega_1$ and hence $t_4 < t_3$. The "echo" we see is not fully understood at present, but we believe that this apparent "echo" may actually be free-induction decay of the ensemble-average $|0\rangle - |1\rangle$ superposition which is created at t_3 as if it were coherent shortly before at $t_4 < t_3$.

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Direct Measurement of Compression of Laser-Imploded Targets Using X-Ray Spectroscopy

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Compression of neon-filled glass microballoons irradiated by a four-beam laser system has been measured directly by Stark broadening, opacity broadening, and spatial profiles of Ne^{+9} x-ray lines. For an 8.6-atm fill pressure and a 0.2-TW, 40-psec laser pulse, the measured compressed neon density was 0.26 g/cm^3 and the product ρR was 2.5×10^{-4} g/cm^2.

The most important parameter in laser-induced fusion experiments, namely the product, ρR, of compressed core density and radius, has been inferred from the dimensions of the region emitting x rays or α particles.[1,2] In this Letter it is shown that spectral profiles of neon x-ray lines from neon-filled targets yield *direct* information on both ρ and ρR. This measurement does not require one to assume that the hot core contains the whole mass of the fill gas; nor does the problem exist of raising an uncertain core radius to the third power. The targets in these experiments were filled with neon only, at either 2.0- or 8.6-atm pressure. However, the same diagnostic methods can apply to a mixture of neon and a thermonuclear fuel; spectra like these obtained here can be expected with only a small amount of neon in future high-ρR experiments.

The experiments were performed on the DELTA four-beam laser system producing power on tar-