Doppler Beats in Superradiance

M. Gross, J. M. Raimond, and S. Haroche
Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure Associé au Centre National de la Recherche Scientifique, 75231 Paris Cédex 05, France
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We report observation of modulations in superradiance, resulting from the beating of light fields emitted by two groups of atoms with different velocities. The beat note is displayed from atomic frequency by the Doppler shift corresponding to the velocity difference (red or blue shift, depending on the direction of superradiant emission). Similarly many-atom beats should occur in superradiance from a mixture of different isotopes. Implications in spectroscopy are considered.

Superradiance has been observed in various recent experiments.[1-5] When the levels involved in the superradiant transition have no structure[4] or when their structure is degenerate or much smaller than the reciprocal of the characteristic superradiance time T_R,[6] a single pulse—sometimes followed by coherent propagation ringings—is detected. When the level structure splittings are of the order of, or larger than, T_R^{-1}, modulations can be observed on the superradiant signal,[5] which have a simple qualitative explanation. The inverted medium is indeed able to amplify two or more different frequencies which beat together during the coherence time of the emission, i.e., during the superradiant pulse duration. The phenomenon is reminiscent of the beating between two lasers and is basically different from the quantum beats observed in ordinary fluorescence. Superradiant beating is indeed a many-atom effect involving interferences between the emissions of phase-matched optical dipoles belonging to different atoms,[7] whereas fluorescence beating is a one-atom effect involving quantum interference between emission amplitudes of different levels belonging to the same atom and prepared in a linear superposition of states. Consequently, superradiant beating can be observed in cases where quantum beats are forbidden such as incoherent preparation of initial substates or structure resolved in the final states of the transition.[5] Another important case where superradiant beats—and not quantum beats—are expected corresponds to initial excitation of two groups of *physically different atoms*, for example different isotopes, or atoms with different velocities. Isotope beats or Doppler beats are then expected, which might have interesting spectroscopic applications. We report in this Letter the first observation of Doppler-shifted beats in superradiance. Preparing two groups of cesium atoms with different velocities in the $7P_{1/2}$ state, we have observed in the superradiant emission towards the $7S_{1/2}$ levels a modulation whose frequency depends on the Doppler shift between the two selected velocities and takes different values for emission in opposite directions.

The Cs energy levels relevant for the experiment are drawn in Fig. 1. As in the previous superradiance beat experiment,[5] the atoms are prepared by a short laser pulse at 4594 Å exciting them to the $7P_{1/2}$ level from one of the hyperfine sublevels $F=3$ or 4 of the $6S_{1/2}$ ground state (we will call g this initial state). The subsequent superradiant emission at 3.1 μm brings the atoms in the two $7S_{1/2}$, $F=3$ and 4 final states. Since hyperfine splitting between these states (2175 MHz) is too large to give rise to observable beats, one can in the following neglect this structure and, for simplicity, consider only one final state, called f. The $7P_{1/2}$ level is also split into two hyperfine substates $F=4$ and $F'=3$ separated by $\nu_{FF'}\simeq 400$ MHz. This structure is buried in the Doppler profile of the pump transition ($\Delta\nu_{\text{Doppler}} \simeq 500$ MHz for each hyperfine component). In order to select two velocity groups in the Maxwellian distribution of the excited state, one can make use of a pump laser having a spectral width $\Delta\nu_L$ much smaller than $\Delta\nu_{\text{Doppler}}$. The atoms are

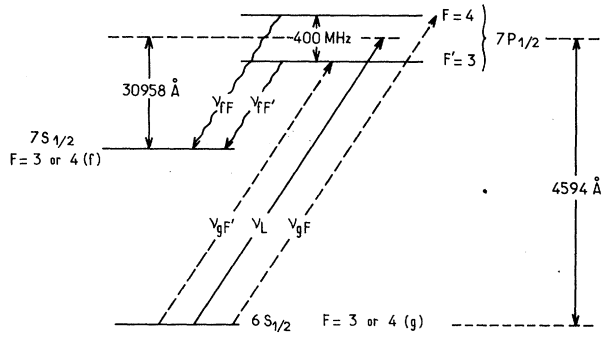

FIG. 1. Scheme of cesium energy levels relevant for the superradiance Doppler beat experiment.

then prepared in the $7P_{1/2}, F$ ($7P_{1/2}, F'$) level if their velocity along the laser beam, V_F ($V_{F'}$) obeys, respectively, the following conditions:

$$\nu_{gF} = \nu_L - k_L V_F, \quad \nu_{gF'} = \nu_L - k_L V_{F'}, \tag{1}$$

where ν_L and k_L are the mean frequency and wave vector of the laser field and ν_{gF} ($\nu_{gF'}$) the optical frequencies of the two pump transitions in the atom rest frame. Two groups of atoms with different velocities are thus excited, which are able to superradiate at frequencies $\nu_{fF} + k_{SR} V_F$ and $\nu_{fF'} + k_{SR} V_{F'}$, respectively, where ν_{fF} and $\nu_{fF'}$ are the rest frame optical frequencies of the superradiant transitions and k_{SR} their mean wave vector. The superradient signal is thus expected to be modulated at the frequency

$$\nu_{\text{beat}} = \nu_{FF'}[1 - k_{SR} k_L^{-1}], \tag{2}$$

which is red shifted with respect to $\nu_{FF'}$ for an emission occurring in the same direction as the laser pulse (forward emission, $k_{SR} k_L^{-1} > 0$) and blue shifted for superradiance in the opposite direction (backward emission, $k_{SR} k_L^{-1} < 0$). The frequency shift, $\pm |k_{SR} k_L^{-1}| \nu_{FF'} = \pm 59$ MHz, corresponds to the Doppler shift between the two velocity groups observed on the infrared transition. The expected beat notes in both directions do not depend on the laser frequency ν_L. However, the beat contrast depends on the respective populations initially excited in levels F and F' and thus changes very rapidly when ν_L is tuned across the Doppler profile of the pump transition. Optimum beat contrast is obtained for a laser frequency ν_L(opt) whose exact value depends on the relative strength of the two pump and two superradiant transitions. In our experiment, we have selected the ground state $|g = 6S_{1/2}, F = 4\rangle$, which corresponds to nearly equal weights for the two pump transitions. In this case, ν_L(opt) is close to $(\nu_{gF} + \nu_{gF'})/2$. In short, the conditions for observation of superradiance Doppler beats can be summarized as follows:

(i) The pump laser should be velocity selective:

$$\Delta\nu_L \ll \Delta\nu_{\text{Doppler}}. \tag{3}$$

(ii) A convenient ratio of atoms in both velocity groups should be excited for good beat contrast:

$$|\nu_L - \nu_L(\text{opt})| \ll \nu_{FF'}. \tag{4}$$

(iii) The superradiant time T_R should be long enough so that at least one beat period could take place during the pulse duration $T_W \sim 10 T_R$ (Ref. 6):

$$T_R \gtrsim (10\nu_{FF'})^{-1}. \tag{5}$$

The experimental setup for observation of the superradiance Doppler beats is sketched in Fig. 2. The exciting laser pulses are produced by a N_2-laser-pumped dye laser (spectral width, about 1500 MHz; pulse repetition rate, 5 pps). The pulses are frequency filtered by a confocal piezo-driven Fabry-Perot etalon, external to the laser cavity (free spectral range, 1500 MHz; finesse, 20). The filtered pulses have a duration $T \simeq 5$ ns, an average peak power of the order of 100 W and a spectral width of 100 MHz, satisfying condition (3). They are sent accross a 10-cm-long cell containing Cs vapor at a temperature of about 50°C, in which they prepare a pencil-shaped amplifying medium with a Fresnel number of about 1. Under these conditions, typical superradiance delays[6] T_D of the order of 20–40 nsec are obtained, which correspond to characteristic superradiant time $T_R \simeq T_D/20 \simeq 1$ or 2 ns, clearly satisfying condition (5). The 3.1-μm superradiant pulses emitted in both forward and backward directions are separated from the laser pulse and from subsequent superradiant emission cascading down from the $7S_{1/2}$ to the $6P$ levels by a filter and a germanium plate acting also as a mirror for the laser beam (see Fig. 2). The two infrared pulses are detected in coincidence with identical fast InSb detectors. Signals from the detectors are amplified and fed into the two channels of a fast R7912 Tektronix transient digitizer. The backward signal is delayed in order to be separated from the forward one on the transient digitizer scope. For observing the Doppler beats, one still has to fulfill condition (4). First, the dye laser itself is tuned on the $6S_{1/2}, F = 4 \rightarrow 7P_{1/2}$ resonance by optimizing the fluorescence from a control Cs cell receiving part of the laser beam

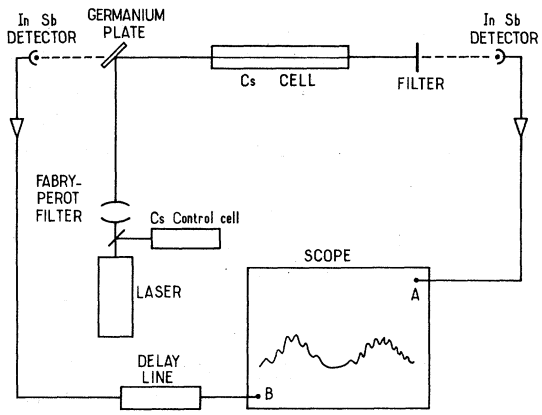

FIG. 2. Scheme of experimental apparatus.

reflected by a plate placed before the Fabry-Perot filter (see Fig. 2). The frequency of the filtered laser pulses is then finely adjusted by sweeping the voltage of the Fabry-Perot peizo-drive until maximum beat contrast is obtained on the signals. When this voltage is offset from its optimum value, first the modulations and then the superradiance signals disappear.

Typical modulated signals obtained in coincidence in both forward and backward directions are shown in Fig. 3(a). The delay of the backward pulse has been substracted, in order to reproduce the exact timing of events ($t=0$ corresponds to the laser pulse maximum). The beat contrast of these single shot signals is good and several beats can be counted. The forward signal appears to have, as expected, a smaller frequency than the backward one. For quantitative frequency determination, a large number of pulses has to be measured. However, because of the delay fluctuations inherent to the superradient emission, no direct averaging technique can be used. One has instead to record independently a large number of forward and backward coincident signals and measure their frequencies, after elimination of the numerous pulses having wrong (too long or too short) superradiant times. A videotape recorder, interfaced to the transient digitizer output is used for fast recording. The histogram of Fig. 3(b) represents the results of beat frequency measurements for a sample of

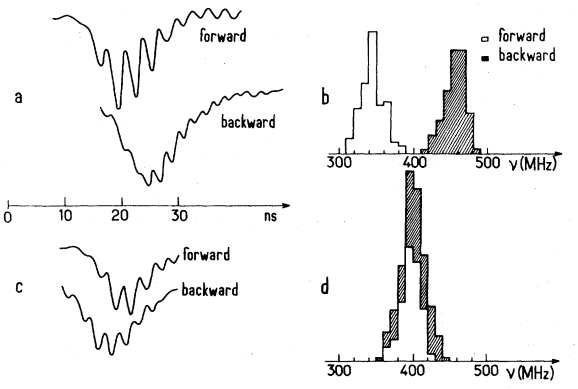

FIG. 3. (a) Recording of Doppler-shifted beats in forward and backward directions following excitation by a narrowband pump pulse. (b) Histogram of beat frequencies in case of narrowband pumping. White and hatched boxes represent forward and backward signals, respectively. (c) Recording of forward and backward signals following excitation by a broadband pump pulse. (d) Histogram of beat frequencies in case of broadband excitation.

100 good laser shots (recording time, about 2 minutes). Forward and backward pulses clearly exhibit different frequency components [$\nu_{for}=344(14)$ MHz and $\nu_{back}=454(16)$ MHz]. The average frequency, $(\nu_{back}+\nu_{for})/2=399(15)$ MHz, is in fair agreement with previous determinations of $\nu_{FF'}$ (377.4 and 401 MHz according to Feiertag, Sahm, and zu Putlitz[8] and Bucka,[9] respectively). The half-frequency difference $(\nu_{back}-\nu_{for})/2=55(15)$ MHz is also in good agreement with the Doppler shift evaluated above. In order to check the importance of condition (3), we have in a control test removed the Fabry-Perot etalon and achieved a broadband excitation of the $7P_{1/2}$ level ($\Delta\nu_L\simeq 1500$ MHz), thus reproducing the conditions under which the beats of Ref. 5 had been observed. Figure 3(c) shows a typical signal recorded in coincidence by both detectors under this condition. Figure 3(d) represents the histogram of frequencies measured in this case on a sample of 100 laser shots. A single frequency component is now observed, with no significant dispersion between forward and backward signals [$\nu_{for}=396(13)$ MHz and $\nu_{back}=398(41)$ MHz]. This result is quite easy to understand. The broadband laser pulses excite now a continuum of velocities in both F and F' levels and the signal contains a large number of Doppler-shifted frequency components around ν_{fF} and $\nu_{fF'}$, resulting in a damped modulation at the average beat frequency $\nu_{FF'}$ in both directions. Comparison between the frequency distributions of Figs. 3(b) and 3(d) shows the critical role played by the spectral profile of the pump pulse in a superradiant beat experiment. The distribution of beat notes clearly reflects the characteristics of the velocity distribution of the initially excited atoms.

This experiment demonstrates the main advantage and limitation of superradiance beat as compared to single-atom quantum-beat spectroscopy. The advantage lies in the possibility of detecting beats coming from *physically different atoms*. In particular, superradiance beats similar to the ones described in this Letter can obviously be used for isotope-shift measurements. The limitation of the technique comes from the sensitivity of these beats to the Doppler effect, which should be carefully controlled in any superradiance beat spectroscopy experiment.

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Study of Atomic Velocities in Molecules Using Nuclear Resonance Photon Scattering

O. Shahal and R. Moreh
Nuclear Research Centre, Negev, Beer Sheva, Israel, and Ben-Gurion University of the Negev, Beer Sheva, Israel
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A strong dependence of the scattering cross section on molecular orientation was observed when 6.324-MeV photons were scattered from ^{15}N in the form of $Na^{15}NO_3$ single crystal. This is caused by the different velocities of the N atom corresponding to the various directions of the *zero-point* molecular vibrations.

We have measured the resonance scattering cross section of 6.324-MeV photons[1,2] from a ^{15}N target in the form of a $Na^{15}NO_3$ single crystal. A strong dependence of the scattering cross section on crystal orientation was observed. This behavior is due to the anisotropic motion of the N atom in the crystal which causes different Doppler broadenings of the nuclear level in the various directions. The results are used to determine the velocity component of the N atom along the photon beam direction. The most important point to be noted here is that the dominant part of this velocity is due to the *zero-point vibration* of the N atom in the various normal modes of vibration of the molecule. The excited vibrational states do not contribute in practice as they are almost unpopulated at room temperature because of the high vibrational energies involved.

The 6.324-MeV level of ^{15}N is photoexcited by a chance overlap (to within ≈ 30 eV) of one of the incident γ lines of the reaction $Cr(n, \gamma)$. The technique involved is described in detail elsewhere.[3]

The Doppler broadening of both the incident γ line and the resonance level in ^{15}N causes an appreciable overlap between the tails of the two lines which gives rise to a relatively high scattering cross section σ_s. σ_s is very sensitive to the Doppler width of the resonance level and hence to the instantaneous velocity of the N atom. In addition, the lifetime of the resonance level ($\tau \sim 10^{-16}$ sec) is much shorter than the period of the molecular vibrational motion ($t \leq 10^{-14}$ sec), and hence the photon is emitted long before any "smearing" of the vibrational velocity takes place. For nuclear levels where $\tau \gg t$, the zero-point vibration can cause almost no Doppler broadening and the effect reported here cannot be observed.

We now consider the motion of the N atom in $NaNO_3$. The NO_3^- molecular ions are planar[4] with their plane perpendicular to the hexagonal c axis of the $NaNO_5$ unit cell. Because of the weak ionic binding between Na^+ and NO_3^-, the kinetic energy of the N atom is due primarily to its internal vibrational motion in NO_3^-. In its ground state, the NO_3^- molecular ion performs zero-point vibrations corresponding to six normal-mode frequencies.[2,4] The first mode does not contribute to the motion of the N atom. In the second mode, ν_2, the N atom vibrates along the

