[10]R. M. Housley, N. E. Erickson, and J. G. Dash, Nucl. Instr. Methods 27, 29 (1964).

[11]D. A. Shirley, M. Kaplan, and P. Axel, Phys. Rev. 123, 816 (1961).

ENHANCED TWO-PROTON EMISSION

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An enhanced two-photon emission has been observed which is due to a strong laser radiation. The intensity of this process is very high when compared with a corresponding spontaneous two-photon emission.[1] Enhancement takes place even though the populations of the emitting atomic or molecular states are not inverted. The laser radiation is coherently amplified and the process provides a method for laser amplification.

Spontaneous two-photon emission is generally a weak process responsible for the decay of metastable states.[1] A continuous distribution of photon pairs is emitted with the energy sum of each pair equal to the separation of the initial and final states of the emitting system (Fig. 1). The contribution of these processes to the decay rate of the emitting state can be obtained by integrating over the distribution of possible photon pairs.[2] This rate is negligibly small compared with that for a cascade of single emissions, if such a process is possible.

A two-photon decay is illustrated in Fig. 1. The energy separation E_3-E_1 is equal to $\hbar\omega_1+\hbar\omega_2$, where ω_1 and ω_2 are the emitted complementary frequencies. Levels E_3 and E_1 have the same parity if the system has inversion symmetry, but the following discussion need not be limited to such systems. The existence of a virtual state E_2 inside the energy interval E_3-E_1 is helpful for two-photon emission, but is not essential.

Physical insight into the important features of the process is obtained if the intensities of the radiation fields are discussed in terms of average photon occupation numbers $\bar{n}_1$ and $\bar{n}_2$ of the radiation modes. For "laser" fields, the average $\bar{n}$ will account only for field modes that are within the angular spread of the beam. This means that, in such fields, the contribution of spontaneous emission to radiation modes outside the laser beam is disregarded.

Let the population densities of the emitting and final states be N_3 and N_1, respectively. The two-photon decay rate for one photon within the frequency region ω_1 and $\omega_1+\Delta\omega$ and a second photon with the complementary frequency is[3]

$$dN_3/dt=-\alpha[(\bar{n}_1+1)(\bar{n}_2+1)N_3-\bar{n}_1\bar{n}_2N_1], \tag{1}$$

where

$$\alpha=\frac{16\pi^3}{3\hbar^2}\bar{g}(\omega_1)\bar{g}(\omega_2)\omega_1\omega_2\Delta\omega^{\frac{1}{3}} \times\left|\sum_{n'}\left(\frac{\mu_{3n'}\mu_{n'1}}{\omega_{n'3}-\omega_2}+\frac{\mu_{3n'}\mu_{n'1}}{\omega_{n'1}+\omega_2}\right)\right|^2. \tag{2}$$

Here $\bar{g}(\omega_1)$ and $\bar{g}(\omega_2)$ are the effective mode densities at the two frequencies where effective mode densities are given by ω^2/π^2c^3 for the case of spontaneous emission. For laser radiation this density contains only the highly excited field modes within the angular aperture of the beam. $\mu_{nn'}$ and $\omega_{nn'}$ are the electric transition dipole and the frequency separation between the levels n and n'.

According to expression (1) the different types of two-photon emissions can be characterized as follows: (a) spontaneous two-photon emission,

$$\bar{n}_1=\bar{n}_2=0;$$

(b) enhanced two-photon emission,

(i) $\bar{n}_1\gg1$, $\bar{n}_2=0$,

or

(ii) $\bar{n}_2\gg1$, $\bar{n}_1=0$;

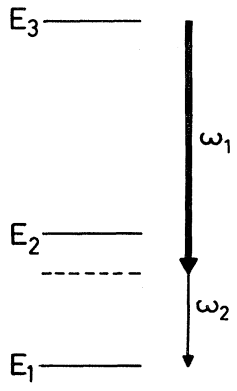

FIG. 1. Level scheme and two-photon emission.

(c) stimulated two-photon transition (emission or absorption),

$$(n_1, n_2) \gg 1.$$

Case (a) is of interest in the decay of metastable states when all the alternative decay rates are very weak. Case (b) is the two-photon decay when the modes at one of the two frequencies are highly excited.

Such a field can be produced by an external laser radiation, which can therefore be used as a primer for an enhanced two-photon process. The decay rate (b)(i) is approximately equal to $-\alpha\bar{n}_1 N_3$ and is therefore proportional to the intensity $\bar{n}_1$ of the laser radiation. There is a preferential emission of the photon pair with frequencies ω_1 and ω_2 as determined by the priming field. The enhanced decay rate can become appreciably faster than that of alternative decay channels even if a cascade of single-photon emissions is permissible.

The two photons are emitted simultaneously, causing an amplification of the priming laser field and a buildup in the occupation $\bar{n}_2$ of the complementary field. If the populations N_3 and N_1 are inverted ($N_3 - N_1 > 0$), an avalanche is possible and $\bar{n}_2$ becomes appreciably larger than unity. The decay rate (1) then assumes the approximate form $dN_3/dt = -\alpha(N_3 - N_1)\bar{n}_1\bar{n}_2$ of a stimulated two-photon emission [case (c)]. This avalanche is reminiscent of the initiation of stimulated Raman emission.

When the population relationship is normal ($N_3 - N_1 < 0$), and if $\bar{n}_2 > 0$, the decay rate (1) is determined by two competing processes: (I) an enhanced two-photon emission rate $-\alpha N_3\bar{n}_1$ which is a spontaneous process with respect to the complementary field at ω_2, and (II) a positive, induced absorption rate $\alpha(N_1 - N_3)\bar{n}_1\bar{n}_2$. The enhanced two-photon emission is thus self-quenched and $\bar{n}_2$ approaches a limiting value $N_3/(N_1 - N_3)$, where N_3 and N_1 are the instantaneous population densities. The saturation of the enhanced process [case (b)] is nevertheless reached when a respectable number of photons has already been emitted at the frequency ω_2. The probability for enhanced two-photon emission before saturation can be written in the form

$$A = \frac{4\pi^2}{3\hbar}\bar{g}(\omega_2)\omega_2\mu_{\text{eff}}^2, \tag{3}$$

where

$$\mu_{\text{eff}}^2 = \frac{4\pi}{\hbar}\bar{g}(\omega_1)\omega_1\Delta\omega\bar{n}_1^{\frac{1}{3}} \times \left|\sum_{n'}\left(\frac{\mu_{3n'}\mu_{n'1}}{\omega_{3n'} - \omega_2} + \frac{\mu_{3n'}\mu_{n'1}}{\omega_{n'1} + \omega_2}\right)\right|^2. \tag{4}$$

In this form the enhanced process is similar to a single-photon spontaneous emission at the complementary frequency ω_2. The effective dipole moment μ_{eff} can be calculated for a specific system and for a known intensity of the laser field. In Eq. (4), effective dipoles of one Debye unit can be obtained with laser intensities of 10^8 W and frequency differences $(\omega_{n'3} - \omega_2)$ of the order of 10^3-10^4 cm^{-1}.

Enhanced two-photon emission has been observed between the 6S and 4S states of potassium atoms [Fig. 2(a)]. The 6S state is excited by the simultaneous absorption of two photons: one at the ruby laser frequency and the other at the Stokes shifted frequency of stimulated Raman emission in nitrobenzene.[4]

The two-photon excitation does not lead to inversion of the 6S and 4S populations. A priming laser field for enhanced two-photon emission from the 6S state is provided by the stimulated atomic Raman scattering of ruby radiation.[4,5] Potassium atoms in the $4P_{3/2}$ excited state have a large resonant Raman scattering cross section for the ruby laser radiation.[5] Intense stimulated Raman radiation at 2720 cm^{-1} is generated by irradiating excited potassium atoms by a giant pulse ruby laser. The $4P_{3/2}$ state is simultaneously populated by the stimulated molecular Raman radiation of nitrobenzene.

The experimental arrangement is similar to that described by Rokni and Yatsiv.[6] Under these conditions the two-photon excitation of the 6S state and the atomic Raman radiation in potassium take place at the same time.

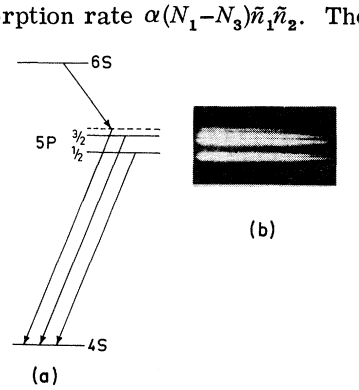

FIG. 2. (a) Energy levels in potassium with resonance and enhanced two-photon emission. (b) Spectrogram of self-reversed resonance lines and of the enhanced emission.

The emission at the complementary frequency ω_2 is located 10 cm^{-1} above the $5P_{3/2}$-$4S$ resonance line in potassium (Fig. 2). For this frequency, the coefficient α in Eq. (1) is close to resonance. The enhanced emission at $\omega_2 = 24\,730$ cm^{-1} satisfies the equation $\hbar(\omega_1+\omega_2) = E_{6S} - E_{4S}$ to within one wave number.

A Polaroid plate showing this emission from the potassium vapor cell is shown in Fig. 2(b). The lines are placed parallel to the corresponding emitting levels in Fig. 2(a). Within each of the $5P$-$4S$ resonance lines there is a black strip which matches exactly the position of the $5P \rightarrow 4S$ resonance emission from a potassium spectral lamp. The resulting doublet structure was observed by Lumpkin et al.[7] and was attributed to wave-function modulation. In our setup there is evidence that at high vapor pressure the emission takes place predominantly at the front of the 1-m-long vapor cell, and the black strips are due to self-reversal by resonant absorption along the cell. The doublet structure in the two lines is not observed when the potassium vapor pressure is sufficiently low.

When nitrobenzene is replaced by bromonaphthalene, the requirement for two-photon excitation of the $6S$ state is not satisfied. Correspondingly, the blue emission at the complementary frequency ω_2 was not observed even though the atomic Raman line appears as usual.[6] This observation is a further evidence for our assignment.

Enhanced two-photon emission is a method for amplifying laser radiation.[8] The virtue of such amplification is that it does not require inversion of population. Enhancement is particularly favorable when the coefficient α in (2) is resonant.

An alternative decay, in which the incident photon ω_1 is absorbed and an anti-Stokes Raman photon $\omega_2 = \omega_1 + (E_3 - E_1)/\hbar$ is simultaneously emitted, can also take place. Such emission is favorable when the "virtual" level E_2 happens to be outside the interval $E_2 - E_1$.

[1]M. Lipeles, R. Novick, and N. Tolk, Phys. Rev. Letters 15, 690 (1965).
[2]J. Shapiro and G. Breit, Phys. Rev. 113, 179 (1959).
[3]M. G. Mayer, Ann. Physik 9, 273 (1931).
[4]Shaul Yatsiv, William G. Wagner, Gerald S. Picus, and Fred J. McClung, Phys. Rev. Letters 15, 614 (1965).
[5]M. Rokni and S. Yatsiv, Phys. Letters 24A, 277 (1967).
[6]M. Rokni and S. Yatsiv, IEEE J. Quantum Electron. 3, 329 (1967).
[7]O. J. Lumpkin et al., Bull. Am. Phys. Soc. 12, 1054 (1967).
[8]P. P. Sorokin and N. Braslau, I.B.M. J. Res. Devlop. 8, 177 (1964).

STRUCTURE IN THE TUNNELING DENSITY OF STATES OF SUPERCONDUCTING La†

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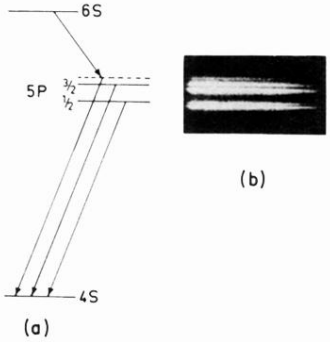
Structure has been observed in the tunneling characteristics of La thin-film diodes which may be attributed to a poorly developed phonon spectrum in the portion of the La film sampled by the experiment.

It has frequently been suggested that a magnetic interaction may be primarily responsible for the superconductivity of some of the transition metals. Such ideas would gain considerable impetus if experimental data were available which were at variance with existing formalisms of superconductivity,[1] and it would help considerably if the experiments were ones which portrayed the electron-phonon interaction in a fairly direct manner so that any variance would be readily recognized.

The electron-tunneling experiment is one of this type,[2] and Wyatt[3] has demonstrated phonon effects in Ta and Nb in this manner. The position of La is less certain. Levinstein, Chirba, and Kunzler[4] have reported seeing phonon effects of amplitudes intermediate between those for Sn and Pb during the course of preliminary point-contact tunneling measurements with La, but Edelstein[5] has reported seeing no such effects with La thin-film tunnel diodes. The lack of sharp phonon effects in thin films may be attributed to lattice disorder, but in view of the results of Chen et al.,[6] who observed phonon effects in amorphous Bi, or of Zavaritskii,[7] who observed phonon effects in Pb films having a deliberately distorted lattice, one would not expect phonon effects to be totally absent in La thin films. This



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