Infrared Laser Action in Sr Vapor Resulting from Optical Pumping and Inelastic Collisions

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Infrared laser action in helium-buffered Sr vapor, resulting from the combined effects of optical pumping and inelastic collisions, has been observed at 2.923, 2.692, and 2.602 μ. A dye laser was used for pumping.

WE have irradiated a column of helium-buffered strontium vapor with the beam from a pulsed dye laser tuned approximately to the Sr $^1S_0 \rightarrow {}^1P_1$ (4607 Å) resonance line and have obtained, besides a relatively weak, but expected, laser emission at 6.45 μ, three comparatively strong infrared laser beams at 2.923, 2.692, and 2.602 μ. These latter emissions correspond to allowed transitions: $^3D_3 \rightarrow {}^3P_2$, $^3D_2 \rightarrow {}^3P_1$, and $^3D_1 \rightarrow {}^3P_0$ (see Fig. 1). An interesting feature is the fact that the emitting triplet states must necessarily be populated by inelastic collisions, the most important ones probably being the following:

$$\mathrm{He}(1^1S)+\mathrm{Sr}(^1P) \rightarrow \mathrm{He}(1^1S)+\mathrm{Sr}(^1D)+\Delta E_1, \quad (1)$$

$$\mathrm{He}(1^1S)+\mathrm{Sr}(^1D) \rightarrow \mathrm{He}(1^1S)+\mathrm{Sr}(^3D)+\Delta E_2. \quad (2)$$

The vapor cell used has been described elsewhere.[1] Briefly stated, its distinguishing feature was that helium gas at roughly one atmosphere was maintained in the cell in order to prevent metal vapor from reaching the end windows.[2,3] A tunable organic dye laser,[4] pumped transversely by the second harmonic beam of a Q-switched ruby laser, irradiated the metal vapor through one of the end windows. With an ethanolic solution of 7-diethylamino-4-Me-coumarin, tunability could be easily achieved over a range of several hundred angstroms in the vicinity of the strontium resonance line. For a given setting of the diffraction grating employed as a reflective element in the laser cavity, the average spectral width of the laser beam was about 30 cm^{-1}. The peak power of the dye laser was roughly 300 kW. (In an auxiliary experiment, described below, a medium-sized (0.2 J), untuned, flashlamp-pumped dye laser containing the same coumarin dye was made to irradiate the cell. This laser, a somewhat simpler version of one previously described,[5] produces pulses of coherent light 2 μsec long with a rise time $\cong$0.5 μsec.)

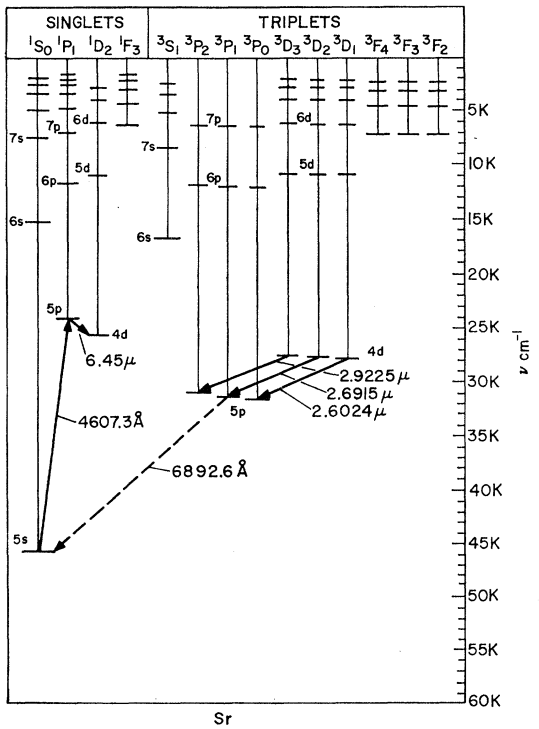

FIG. 1. Energy levels of Sr.

The infrared laser beams produced in the Sr vapor were observed with a room-temperature operated InSb detector whose response time was 50 nsec. The exit window of the vapor cell was a polished Si wafer. This was to allow the expected 6.45-μ radiation to pass. A pulsed beam at this wavelength was indeed detected, as already noted, but its peak intensity was quite small, only about 0.4 kW. By contrast, the sum of the peak intensities of the three simultaneously occurring $^3D_i \rightarrow {}^3P_{i-1}$ laser beams was at least 4 kW. The relative intensities of the three lasing components $^3D_i \rightarrow {}^3P_{i-1}$ were roughly in the ratio 5:2:1 for $i=3$, 2, and 1, respectively. No laser beams were observed to occur at 3.067, 2.736, or 3.011 μ. These wavelengths correspond to the transitions $^3D_1 \rightarrow {}^3P_2$, $^3D_1 \rightarrow {}^3P_1$, and $^3D_2 \rightarrow {}^3P_2$ which, although allowed, have considerably smaller spontaneous emission probabilities than the transitions $^3D_i \rightarrow {}^3P_{i-1}$, at least in calcium.[6] To determine the various wavelengths around 3 μ a 0.5-m Jarrell-Ash monochromator was used.

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As summarized, for example, in Bennett's paper,[7] the cross sections for reactions such as (1) and (2) have the following properties: They are very large ($\cong 10^{-14}$–10^{-13} cm^2) when the energy discrepancy is small and total spin is conserved; they are still quite appreciable ($\cong 10^{-16}$–10^{-15} cm^2) between states having energy differences of several kT, even when Wigner's spin-conservation rule is apparently violated; they are negligible ($\sim 10^{-20}$ cm^2) when the energy difference exceeds, say, 0.5 eV (4000 cm^{-1}) in the case that the atoms have thermal velocities corresponding to $T=300°$K. In our experiment the temperature of the vapor is more like 1100°K, so that, for example, the energy difference between states ^1P and ^1D is only $\cong 2\ kT$ and that between the ^1D and ^3D states is only slightly greater, about 2.5 kT. It would thus appear reasonable to assume a value of 10^{-16} cm^2 for the cross sections of reactions (1) and (2).

Assuming a value 2.4×10^5 cm/sec for the mean thermal velocity of the helium atoms and a He density of 8×10^{18}/cm^3, one easily calculates that excited Sr atoms should undergo reactions (1) and (2) at a rate $\sim 2\times 10^8$ sec^{-1}. Thus in $\sim$5-nsec state, ^1D should start to be appreciably filled as a result of collisions of He atoms with Sr atoms in the ^1P state. This is enough nonradiative leakage to cause serious diminution of the 6.45-μ laser pulse, as the emitting state—the same ^1P state—is pumped in our experiment by a pulse that rises in 10 nsec. A reasonable explanation for the observed low intensity of the 6.45-μ laser pulse is thus afforded. The spontaneous lifetime, incidentally, of the transition in Ca analogous to the 6.45-μ Sr line is about 30 μsec.[6]

State ^1D is completely stable, as regards radiative decay, so that collisional transfer of energy from the ^1D to the ^3D states should be quite efficient in the sense of conserving numbers of excited particles. It is assumed that no significant loss of particles occurs through collisions which deactivate ^1D directly to ^3P ($\Delta E\cong 7.5kT$). Evidently this is so, since even collisional deactivation of the ^3D-to-^3P states ($\Delta E\cong 5kT$) is very slow, as we now show.

The strongest evidence for the last statement is the fact that relatively intense lasing was also observed in the $^3D_i \rightarrow {}^3P_{i-1}$ transitions when a flashlamp-pumped dye laser, the auxiliary source mentioned earlier, was used for excitation. Since the rise time of the output pulse is here $\cong 0.5$ μsec, the ^3D-to-^3P collisional relaxation time must be at least of this order. It thus appears that increasing the energy difference from $\Delta E\cong 2kT$ to $\Delta E\cong 5kT$ in what should be basically the same kind of transition (^1P to ^1D versus ^3P to ^3D) increases the collisional deactivation time by at least two orders of magnitude. The spontaneous emission decay time for the $^3D_i \rightarrow {}^3P_{i-1}$ transitions should be about 400 nsec, by analogy with calcium.[6]

As a sort of corollary, it follows that it should be possible to achieve laser action in the partially forbidden ($\tau\cong 20$ μsec [8]) $^3P_1 \rightarrow {}^1S_0$ resonance line at 6892.6 Å. To invert this transition one would have to pump the 4607.3-Å line with a somewhat larger version of the auxiliary dye laser used in this experiment. If the 6892-Å laser can be realized, it would be possible to Q-switch it.

We would like to thank a colleague, Dr. R. Hodgson, for helpful discussions regarding collisional transfer.

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