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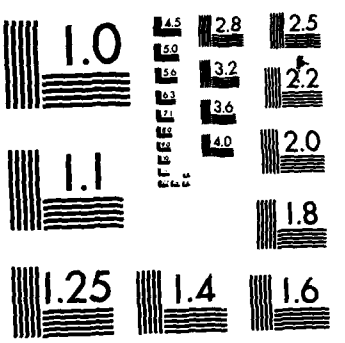

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uous-wave lasing; vibrational transition; population inversion; optical ; overtone; tunable; color center laser; energy budget; solid-state nal laser; color center; fluorescence; vibrational energy exchange; ransfer; inhomogeneous; Manley-Rowe

ABSTRACT (Continue on reverse side if necessary and identify by block number)

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Using pulsed fluorescence techniques and numerical simulations, we find that due to vibrational energy exchange between pairs of CN^- ions in the v = 1 lower laser level, "waste" pump energy is recycled to the v = 2 upper level. Ideally the efficiency of this recycling scheme approaches unity, but due to energy transfer into higher vibrational levels, and because of spectral diffusion within the inhomogeneously broadened CN^- profile, the observed recycling efficiency is reduced. To our knowledge this is the first experiment demonstrating defeat of the Manley-Rowe limit. Keywords: [illegible]

Development of Solid State Vibrational Lasers

4/1/84 - 3/31/85

FINAL REPORT

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August 1, 1985

U. S. Army Research Office

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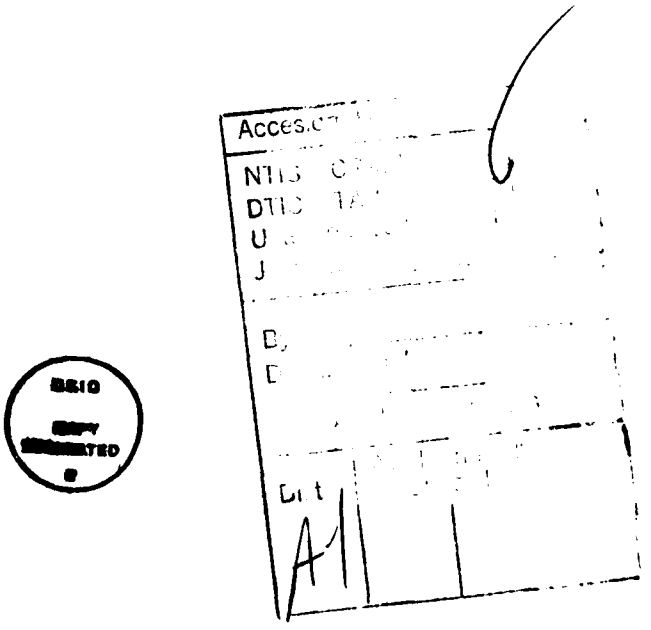
Cornell University

TABLE OF CONTENTS



I. Summary

Continuous-wave lasing in the 5 μm wavelength region has been obtained on the 2→1 vibrational transition of impurity CN^- ions in KBr. Population inversion is produced by optical pumping of the weakly allowed first-overtone level of the molecule with a tunable $(F_2^+)_A$ color center laser.

We have investigated the energy budget of this solid-state vibrational laser when the v = 2 upper laser level is pumped with a CW color center laser. Using pulsed fluorescence techniques and numerical simulations, we find that due to vibrational energy exchange between pairs of CN^- ions in the v = 1 lower laser level, "waste" pump energy is recycled to the v = 2 upper level. Ideally the efficiency of this recycling scheme approaches unity, but due to energy transfer into higher vibrational levels, and because of spectral diffusion within the inhomogeneously broadened CN^- profile, the observed recycling efficiency is reduced. To our knowledge this is the first experiment demonstrating defeat of the Manley-Rowe limit.

II. Statement of the Problem

With the discovery of vibrational fluorescence from CN^- ions embedded in KCl host matrices[1] came the first possibility for the construction of a solid-state laser based on purely vibrational transitions. This possibility was recently realized in KBr crystals doped with 0.5 mol % KCN when optical pumping between the v = 0 and v = 1 levels produced lasing between the v = 2 and v = 1 vibrational levels of CN^- in a pulsed mode.[2] The crucial mechanism leading to a population inversion in this high concentration system is the vibrational energy exchange between proximate CN^- ions in their v = 1 state. The result of this energy exchange (V-V transfer is the rapid depletion of the v = 1 lower laser level population in favor of the v = 2 upper laser level and the v = 0 ground state.

Although vibrational energy exchange between diatomic molecules in IR gas lasers has been well studied, the observation of laser action on a vibrational transition of CN^- ions embedded in KBr[2] raises new questions about the role of V-V transfer processes in this different context. In addition reliance on this interesting and unusual pumping scheme imposes two important limitations on the laser: The V-V transfer process limits the device only to pulsed operation when pumped on the 0→1 transition, and second, at increased temperature V-V transfer depopulates the v = 2 state in favor of the v = 1 state, restricting laser operation to temperatures below 4 K.

In this final report we describe our successful attempt to overcome these limitations. A tunable $(F_2^+)_A$ color-center laser is used to pump directly the weakly allowed 0→2 vibrational transition of the CN^- ion at 2.4 μm in KBr crystals doped with 0.5 and 0.05 mol % KCN. Continuous-wave lasing is observed to occur on the 2→1 transition at 4.9 μm for both crystals.

III. Experimental Investigation

The doped KBr crystals used in these measurements are prepared by the method described in Ref. 2. Resonant cavities are fabricated by evaporating reflective films directly onto the 1 mm x 1 mm end faces of cleaved crystals of different lengths. For 0.5% concentration a 5-mm long cavity is formed with a λ/4 (at 4.9 μm) germanium layer on one end face and a gold layer on the other. Efficient input coupling of the 2.4-μm pump light is possible through the germanium layer, which has an antireflection coating at the pump wavelength but as a 70% reflector at the lasing wavelength. For the 0.05% concentration, gold films are used on both ends of a 14-mm long cavity, with input coupling provided by a 0.2 mm hole in one of the films. For ease of optical alignment on both crystals, output coupling of a few percent is provided by a small hole in the gold end reflector opposite the input coupler.

The KBr crystal cavities are attached with wire to the bottom of a stage suspended in a liquid helium bath. The temperature of the laser cavities is variable down to 1.7 K. The color-center laser beam is focused by a 100 mm lens through two cryostat windows into the end mirror of the KBr cavity. No attempt was made to mode match the pump spot with the cavity, as the plane-parallel structure of the KBr lasers certainly permitted operation in numerous transverse modes. The output coupling hole on the opposite end of the laser is imaged either directly or through a 0.5 m monochromator onto a 100 kHz bandwidth InSb detector. Lasing is unmistakably detected by a sudden off-scale increase of the detected signal on a monitoring oscilloscope and by the rapid rise and fall times of the signal compared with the radiative lifetime (25 msec).

The color-center pump laser is based on the $(F_2^+)_A$ center in lithium-doped KCl. The necessary pump wavelength (2.42 μm) is near the extreme end of the tuning range of the color-center crystal, so output power is limited to a continuous-wave value of 13 mW. Laser construction is similar to that described in Ref. 3. Coarse tuning is achieved by placing a sapphire prism in the output arm of the color-center laser cavity and rotating the output coupler. This construction also separates residual Nd:YAG laser pump radiation from the color-center output, thus reducing the subsequent heat load on the liquid-helium cryostat. Fine tuning the color-center laser across the absorption profile of the 0→2 transition of the CN^- ions is accomplished by angle tuning a 30% reflective 1 mm interacavity étalon. The final linewidth of this color center laser is less than 1 GHz, which coresponds to two or three active longitudinal modes of the cavity.

A level diagram depicting the overtone pumping scheme is shown in Fig. 1. From Fourier-transform spectrometer measurements the inhomogeneous linewidth of the 0→2 transition is 0.5 cm^{-1}, and the absorption coefficient at line center is 2.1 cm^{-1} for the KBr sample containing 0.5 mol KCN at 1.7 K. The strength of this transition is roughly 1% that of the fundamental; however, it is still strong enough to absorb most of the pump energy in a 1-cm-long crystal. The gold mirrors on both ends of the lightly doped crystal help to trap the pump radiation, increasing the effective absorption.

Figure 2(a) shows the output from the 0.5 mol % cavity at 1.7 K (upper trace) along with the chopped output of the 13-mW pump laser. Note the brief delay following the leading edge of the pump pulse before oscillation begins. This delay is due to the necessary buildup of gain to reach theshold. The trailing edge of the output pulse closely follows the pump. This observation suggested the possibility of continuous-wave laser operation, and this was confirmed for both the 0.5 and 0.5% cavities. A typical trace is shown in Fig. 2(b). The output power for both cavities was similar and was estimated to be a few microwatts, although the output power through the germanium input coupler of the 0.5% cavity was probably several times larger.

One expected characteristic of the laser output for such a relatively long-lived upper state is relaxation oscillations. These are visible in the traces of the output power.

The temperature dependence of the gain in both cavities is found to be very similar to that of the pulse laser with no lasing occurring above 7 K.

IV. Discussion and Conclusions

The fact that cw operation of this device is observed is perhaps at first surprising, since a bottleneck is expected at the lower laser level due

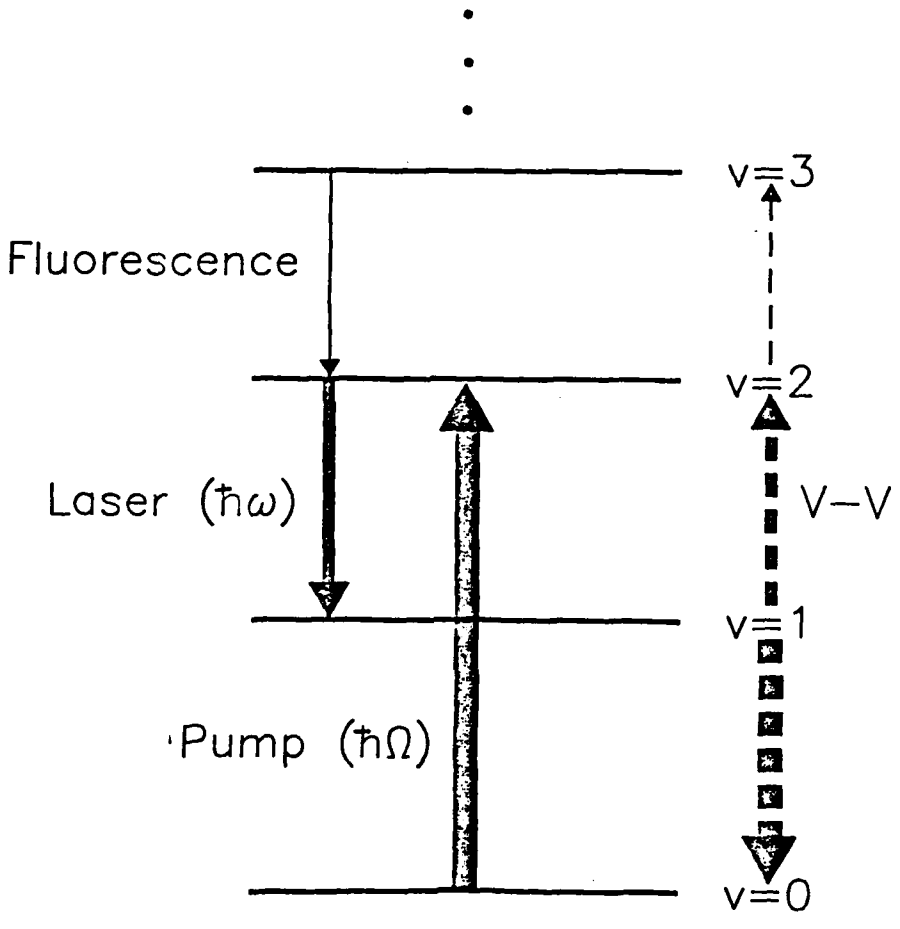


Figure 1: Energy level diagram of the anharmonic vibrational manifold of CN^- showing schematically the repopulating effects of energy exchange. Optical transitions are indicated as solid arrows, while dashed arrows show nonradiative transitions.

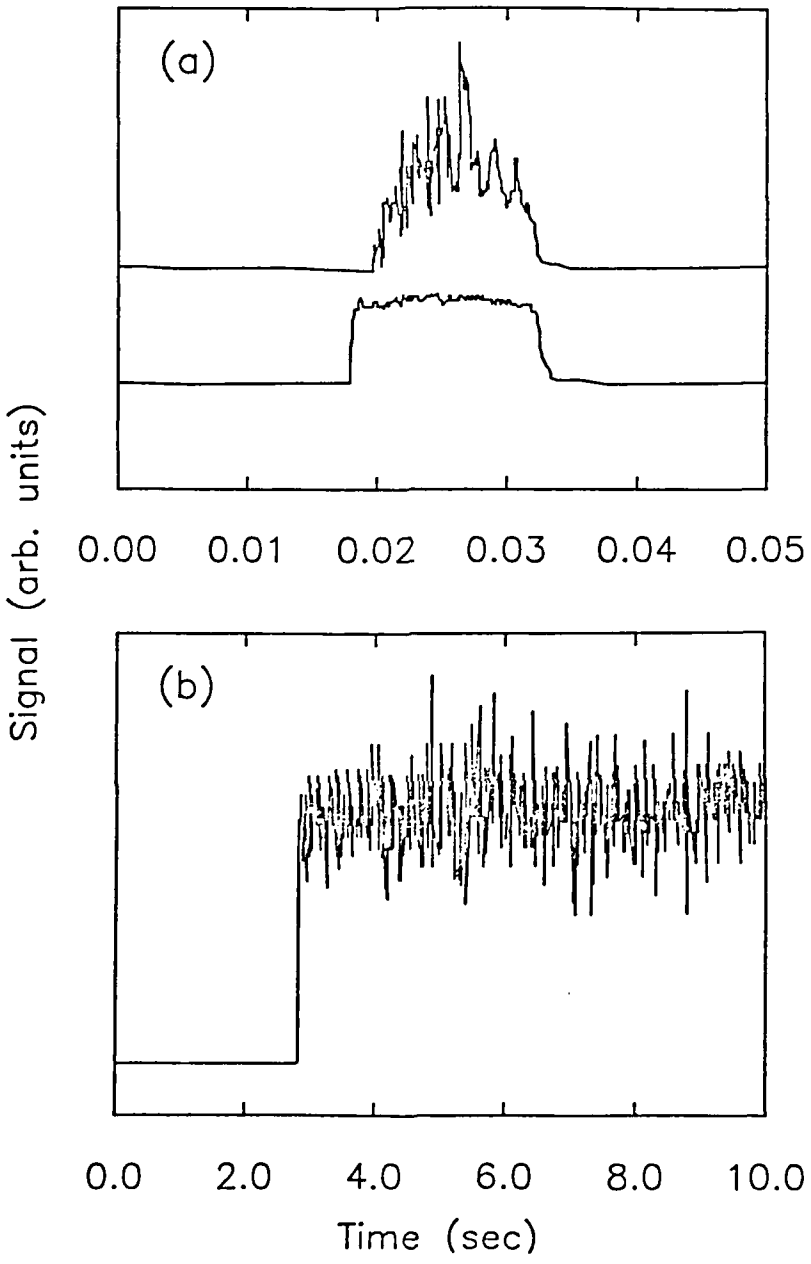


Figure 2: (a) Lasing at 2054 cm^{-1} from a 0.5% KCN-doped crystal at 1.7 K (upper trace) versus the chopped pump pulse at 4130 cm^{-1}. (b) Continuous-wave lasing of a 0.05% sample. Peak output power for both types of crystal is estimated at a few microwatts.

to the long radiative lifetime (~50 msec) of the v = 1 state. However, rapid depletion of the v = 1 state population by V-V transfer process (1) as well as by the general exchange reaction

$$CN^-(1) + CN^-(v) \rightarrow CN^-(0) + CN^-(v+1) + \Delta E \qquad (1)$$

reduces the effective lifetime of the lower laser level to <1 msec.

Direct evidence for the lifetime shortening effect of V-V transfer on the v = 1 state is shown in Fig. 3. An initial population in the v = 1 state is produced with a short pulse of doubled CO_2 laser radiation, and the subsequent fluorescence signals indicate the time evolution of the vibrational populations. Note the rapid fall in signal from the 1 → 0 transition and the corresponding fast rise of the 2 → 1 fluorescence signal.

The quenching of the laser gain is due to the thermally induced V-V transfer reaction[2,4]

$$CN^-(2) + CN^-(0) + 25\ cm^{-1} \rightarrow 2CN^-(1), \qquad (2)$$

which rapidly depletes the upper laser level. On the other hand, the spontaneous reverse process, which dominates below 4 K, acts to repopulate the n = 2 level once the 2 → 1 transition becomes saturated. Indeed, only weak fluorescence from the 1 → 0 transition is observed at low temperatures. Below 4 K, this transfer back to the v = 2 state reduces to less than 1 msec the effective lifetime of the lower laser level.[5]

An important consequence of overtone pumping is the following: Pump energy delivered to the v = 1 level by gain saturation of the 2→1 transition is partly recycled to the upper laser level via process (2). A more complete

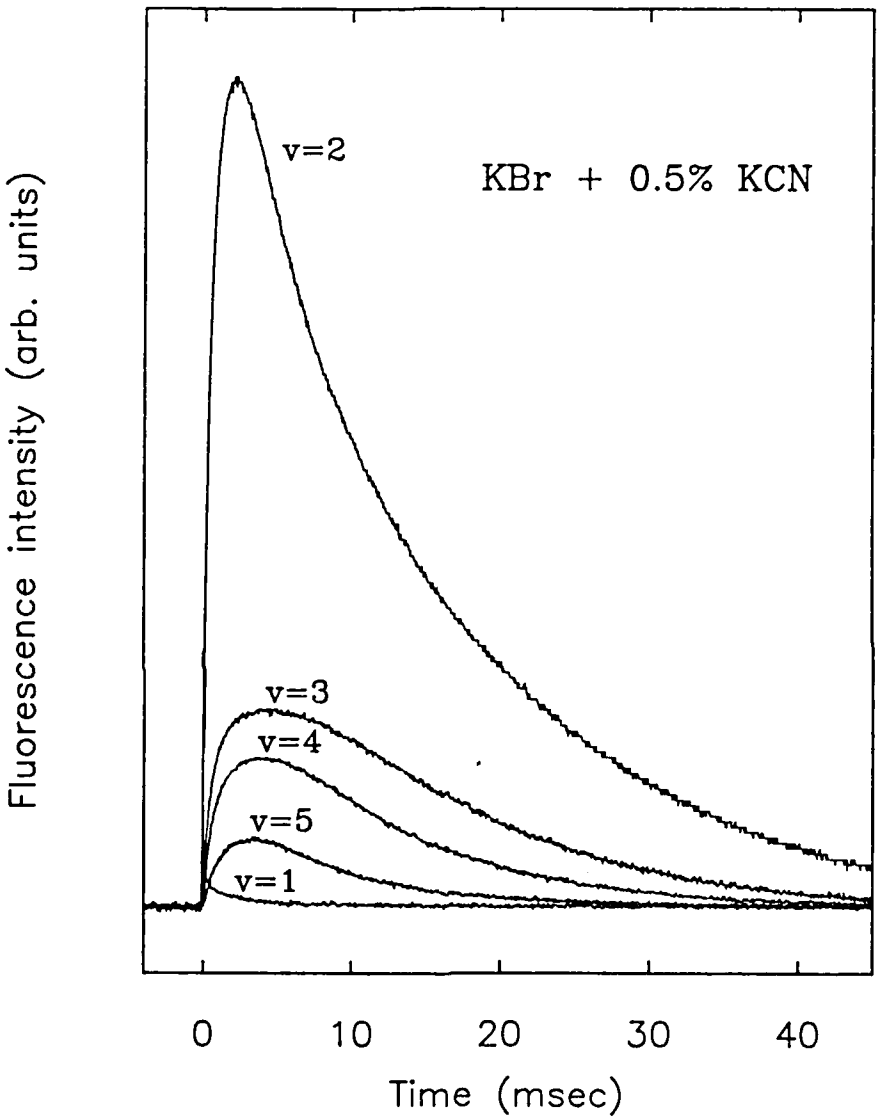


Figure 3: Fluorescence signals from the first five vibrational transitions of CN^- following pulsed pumping of the v = 1 state. V-V transfer rapidly depletes the v = 1 level.

accounting of the budget in these circumstances is illustrated in the level diagram of Fig. 1 which includes the influence of process (1). Normally, the efficiency of an optically pumped three-level laser is constrained by the Manley-Rowe limit $\varepsilon = \omega/\Omega$, where Ω is the pump frequency and ω is the output frequency. In the KRr:CN^- system, however, V-V transfer bypasses the Manley-Rowe limit, which in an ideal case would lead to efficiencies exceeding 50%. Although our observed overall efficiency is much less than this value due to external and internal effects unrelated to V-V transfer, the recycling process does play an important role. These other operating efficiencies aside, the KBr:CN^- laser is the first experimental system to our knowledge to defeat the Manley-Rowe limit.

The results presented here demonstrate the feasibility of an alternative pumping scheme for the KBr:CN^- laser that should lead to higher-temperature operation with broad tunability. Although the effect of thermal quenching of the gain through V-V transfer is still dominant at 0.05 mol % CN^-, reducing the concentration even further will eventually decouple the CN^- dipoles, permitting inversion above 4 K. Additionally, the rapidly increasing absorption linewidth of the ions resulting from the thermal occupation of hindered rotor levels[6] (FWHM of 50 cm^{-1} at 50 K), combined with the measured high fluorescence quantum efficiency at 80 K (Ref. 7) and even at room temperature,[8] gives promising prospects for more convenient and useful operation of this device in the 5-μm spectral region.

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VII. Scientific Personnel

R. Tkach, K. Blum, A. J. Sievers

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