Characterization of Spontaneous and Stimulated Emission from Praseodymium (Pr^{3+}) Ions Doped into a Silica-Based Monomode Optical Fiber

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Abstract—**The fluorescence spectrum of praseodymium in a silica host has been investigated through the direct excitation of the $^3P_0$, $^1D_2$, and $^1G_4$ absorption bands, which occur at around 488, 590, and 974 nm, respectively. The observed spontaneous emission consists of four main bands with peaks at 633, 707, 888, and 1080 nm. In each case the level principally responsible for these emissions was found to be $^1D_2$. Stimulated emission has been seen for the first time at 888 nm for a threshold of 10 mW absorbed power. In addition, the lasing characteristics at 1080 nm have been investigated.**

Introduction

TRIVALENT praseodymium is an interesting laser activator because its energy level spectrum contains an exceptionally large number of metastable multiplets ($^3P_{2,1,0}$, $^1D_2$, $^1G_4$) from which laser action has been demonstrated in crystalline hosts at various wavelengths from the visible [1], to the near infrared [2]. The assignment of transitions to the broad praseodymium fluorescence bands observed in glass hosts is correspondingly difficult in the cases where a band may be composed of spectrally overlapping contributions from more than one initial level. Laser action of praseodymium-activated bulk glass has not to our knowledge been reported previously in the literature, probably because praseodymium has fewer suitable absorptions for flashlamp radiation compared, for example, to neodymium. Thus thresholds for flashlamp-pumped lasing in Pr^{3+} : crystal systems are high, while in glasses the increase in the linewidth by a factor of 10 or more raises the threshold to an impractical level.

More efficient use of pump energy is achieved with a resonant, longitudinal, laser-pumping scheme, particularly when the gain medium is in the form of an optical fiber waveguide rather than a bulk glass. With both pump and laser radiation fields guided in modes of small cross-sectional area the threshold for oscillation is reduced in proportion, and the possibility of achieving threshold on transitions with low radiative quantum efficiency or small branching ratios arises.

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Laser action in a praseodymium-doped silica fiber has been reported by Reekie *et al.* [3], who observed emission at 1080 nm when pumping at 590 nm with radiation from a rhodamine 6*G* dye laser. The authors suggested that the $^1G_4$–$^3H_4$ (ground multiplet) transition was responsible, as in for example the crystalline Pr^{3+} : $CaWO_4$ system which has been shown to lase at 1047 nm [2]. However, the paper by Ainslie *et al.* [4] points out that the emission band at $\simeq 1$ μm will contain overlapping contributions from the $^1D_2$–$^3F_{4,3}$ and $^1G_4$–$^3H_4$ transitions.

In this paper we describe the laser performance of a praseodymium-doped silica fiber at 1080 nm, and also on a transition at 888 nm for which laser action has not hitherto been reported. Comparison of fluorescence spectra excited using different pump wavelengths shows that the upper level of not only the 888 nm emission but also the 1080 nm emission is most likely to be in the $^1D_2$ multiplet. Indeed for praseodymium ions in silica glass, emission from the $^1D_2$ multiplet appears to dominate the fluorescence spectrum and to be principally responsible for the 633 and 707 nm bands which Ainslie and co-workers attribute to the $^3P_0$ multiplet [4].

The spectroscopic and laser performance data reported here indicate that in this fiber the $^3P_0$ level suffers a fast nonradiative decay which does not transfer population to the $^1D_2$ multiplet, thus preventing the realization of visible laser operation via Ar^+ ion pumping, as had been previously proposed [3].

Fluorescence Spectroscopy

All of the experiments used a silica-based fiber prepared by the modified chemical vapor deposition (MCVD) technique [5]. A comparison of the measured absorption coefficient, with data for absorption in a silicate glass from Smith and Cohen [6], yielded an upper bound for the praseodymium ion concentration of $\simeq 10^{25}/m^3$. The fiber is further characterized by a core diameter of 2.5 μm and an LP_{11} mode cut-off wavelength of 890 nm.

The absorption spectrum of this fiber contains three bands in the 400–1100 nm region which correspond to the $^3H_4$–$^3P_{0,1,2}$, $^1D_2$, and $^1G_4$ transitions (see energy level dia-

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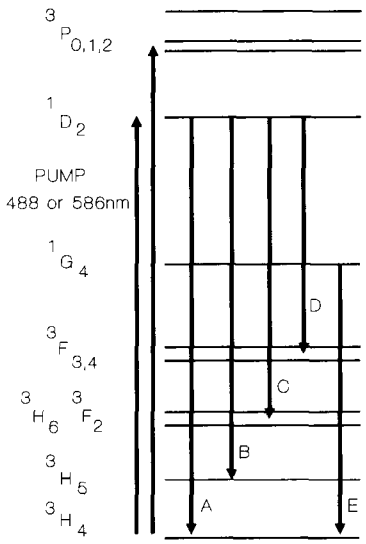

Fig. 1. An energy level diagram of praseodymium (not to scale).

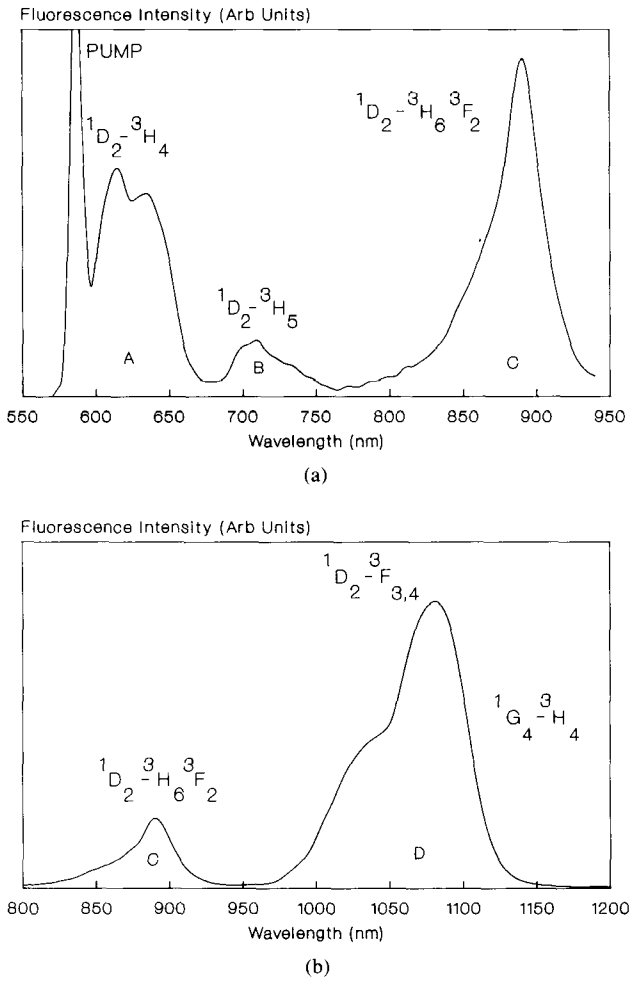

Fig. 2. (a) The fluorescence spectrum of Pr^{3+} in fused silica 550–950 nm, recorded in side-light, under excitation from an Rh6*G* dye laser at 590 nm. The strong peak at 590 nm is due to scattered pump light. (b) The fluorescence spectrum of Pr^{3+} in fused silica 800–1200 nm, recorded in side-light, under excitation from an Rh6*G* dye laser at 590 nm.

gram in Fig. 1) at 450–490, 580–590, and 970–980 nm, respectively [4].

Fig. 2(a) and (b) show the four fluorescence bands excited by pumping the fiber with an Rh6*G* dye laser tuned into resonance with the $^1D_2$ absorption band at around 590 nm. The experimental arrangement used to measure a "side-light" spectrum is shown in Fig. 3. Unguided fluorescence escaping radially out of the fiber core is collected using a slit-to-slit fiber optic bundle, analyzed by a 0.25 m grating monochromator, and detected using a large area silicon photodiode [Fig. 2(a)] or a room temperature germanium photodiode [Fig. 2(b)]. Since the optical attenuation of the fiber core in a radial direction is extremely small ($<10^{-5}$ dB) even near the peak of an absorption band the fluorescence spectrum is not distorted by ground state reabsorption. No correction has been made for the spectral response of the monochromator and detector. The fluorescence bands *A* (633 nm), *B* (707 nm), and *C* (888 nm) can be assigned unambiguously as being from the $^1D_2$–$^3H_4$, $^1D_2$–$^3H_5$, and $^1D_2$–$^3H_6$, $^3F_2$ transitions, respectively. The remaining band *D* (1080 nm) must be due to one or both of the $^1D_2$–$^3F_{3,4}$, and $^1G_4$–$^3H_4$, transitions. Emission band *A* ($^1D_2$–$^3H_4$) shows two partially-resolved peaks at 610 and 630 nm. Since the emissions presumably originate from the lowest energy Stark level of $^1D_2$ these peaks must represent Stark levels in the $^3H_4$ ground multiplet. The large Stokes shift of the 630 nm peak shows that the $^3H_4$ multiplet is split by some 1300 cm^{-1}.

Measurements of fluorescence lifetime were made using either a photomultiplier or a silicon photodiode with a digitizer to monitor the decay of the fluorescence following mechanical interruption of the pump beam. The time resolution for the measurements is less than the quoted uncertainty. All four fluorescence bands exhibited approximately exponential decays with the same $1/e$ time of 120 $\pm$ 10 μs, which must therefore be the lifetime of the metastable level in the $^1D_2$ multiplet. This evidence suggests that the 1080 nm fluorescence band is dominated by the $^1D_2$–$^3F_{3,4}$ transitions.

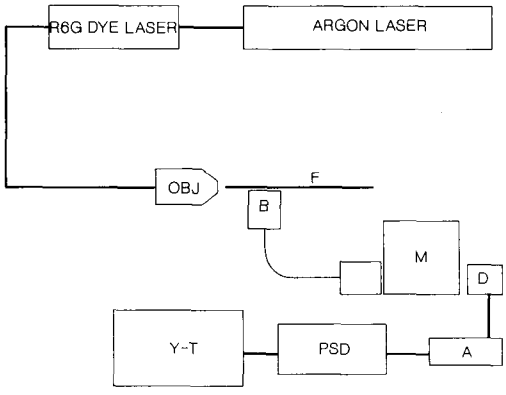

Fig. 3. Experimental configuration for the monitoring of side scattered emission from a longitudinally pumped fiber. *F* = sample fiber; *B* = slit-to-slit fiber optic bundle; *M* = monochromator; *D* = detector; *A* = amplifier; PSD = lock-in-amplifier; *Y-T* = chart recorder.

To resolve the assignment of the 1080 nm emission the fiber was pumped at 974 nm, almost on resonance with

the $^1G_4$ level, with several milliwatts of superfluorescent power generated by an ytterbium-doped silica fiber source [7]. As described in [7], besides emission at 974 nm this source also produces strong superfluorescent output at 1035 nm, which was filtered out using a 0.25 m monochromator prior to launch into the fiber. Fig. 4 shows the spectrum of guided fluorescence emerging from the end of a 1.3 m length of fiber under 974 nm excitation. The single emission band at 1102 nm can only be due to the $^1G_4$–$^3H_4$ transition. The short wavelength side of this emission band may show the effect of reabsorption by the $^3H_4$ ground multiplet. Since the fluorescence is weak, the radially-emitted portion could not be detected with adequate signal-to-noise ratio. The 1080 nm fluorescence is therefore shown to be a superposition of $^1D_2$–$^3F_3$ (1030 nm), $^1D_2$–$^3F_4$ (1080 nm), and $^1G_4$–$^3H_4$ (1102 nm), which is hidden in the long wavelength tail of the band. As with the $^1D_2$–$^3H_4$ transition, the emission shows a large Stokes shift relative to the corresponding absorption which peaks at 970 nm, confirming that the ground state has a Stark splitting of $\simeq 1200$ cm^{-1}.

Fig. 5 shows the side-light spectrum excited when the fiber is pumped with 488 nm radiation from an argon ion laser into the $^3P_0$ absorption band. A strong scattered pump light feature appears at 488 nm, partially masking an emission band peaking at 510 nm which must be due to the $^3P_0$–$^3H_4$ transition. A comparison of the appearance of emission band *A* in Fig. 5 and in Fig. 4(a) where the $^1D_2$ multiplet is pumped directly shows that an extra shoulder at 650 nm appears under $^3P_0$ pumping. This can be attributed to the $^3P_0$–$^3H_6$, $^3F_2$ transitions which make, however, a relatively weak contribution to the total red fluorescence. These are the only two emissions from $^3P_0$ which appear clearly in the spectrum. The $^3P_0$–$^3H_5$ transition which would peak at $\simeq 550$ nm is strongly forbidden; the $^3P_0$–$^3F_4$ emission at $\simeq 747$ nm may be present under the tail of the $^1D_2$–$^3H_5$ band, but if present is very weak.

Of the three strong emission bands *A*, *C*, and *D* which are potentially interesting as tunable laser transitions, *A* and *D* overlap spectrally with the praseodymium absorption bands. The effect of ground-state reabsorption is illustrated in Fig. 6(a) and (b), which show the spectrum of guided fluorescence emitted from the end of a 1.3 m length of fiber pumped by $\simeq 200$ mW of 590 nm radiation. In emission band *A* ($^1D_2$–$^3H_4$) the 610 nm peak (which is strongly evident in the side-light spectrum of Fig. 2) has been reabsorbed, however the large $^3H_4$ Stark splitting means that the 630 nm peak is essentially unaffected. The $^1D_2$–$^3H_4$ transition therefore changes character from three-level to four-level between the short wavelength and long wavelength tails. Bands *B* and *C* are remote from absorption wavelengths and as expected show no change of shape between side-light and end-light. In emission band *D* the shoulder at $\simeq 1030$ nm ($^1D_2$–$^3F_3$) is suppressed by reabsorption in the long wavelength tail of the $^3H_4$–$^1G_4$ transition, but the strong 1080 nm peak is unaffected. The effect of ground state reabsorption makes it difficult to predict the likely tuning range of laser emission on transitions *A* and *D* that could actually be achieved.

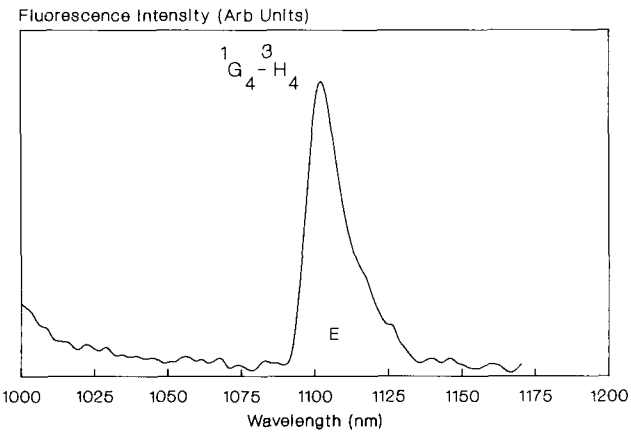

Fig. 4. The fluorescence spectrum of Pr^{3+} in fused silica 1000–1200 nm, recorded in endlight, under excitation from a superfluorescent ytterbium fiber source at 974 nm.

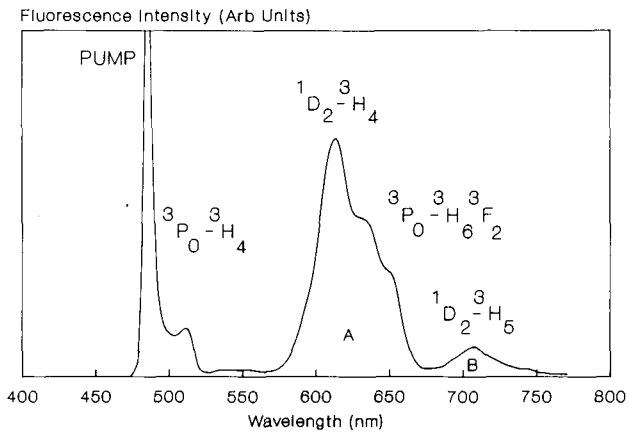

Fig. 5. The fluorescence spectrum of Pr^{3+} in fused silica 400–800 nm, recorded in side-light, under excitation from an argon ion laser at 488 nm.

Laser Performance

To investigate the laser performance of these transitions a cavity was formed by butting dielectric mirrors to both cleaved end faces of a 1.3 m length of fiber. Pump radiation at 590 nm from a rhodamine 6*G* dye laser was launched into the fiber through one mirror using an 18× microscope objective.

With a pair of mirrors selected to have reflectivities of >99.5 and $\simeq 98$ percent at around 1080 nm the threshold for stimulated emission was $\simeq 2$ mW absorbed power. The overall cavity losses (for a single pass) with these mirrors were measured to be 2 percent using a relaxation oscillation technique [8]. Since the cavity contained no spectrally-selective elements, lasing occurred at several wavelengths over a range of 10 nm centered at 1080 nm. Fig. 7 shows the dependence of output laser power on absorbed pump power, observed when an output coupler of nominally 14 percent transmission was substituted for the

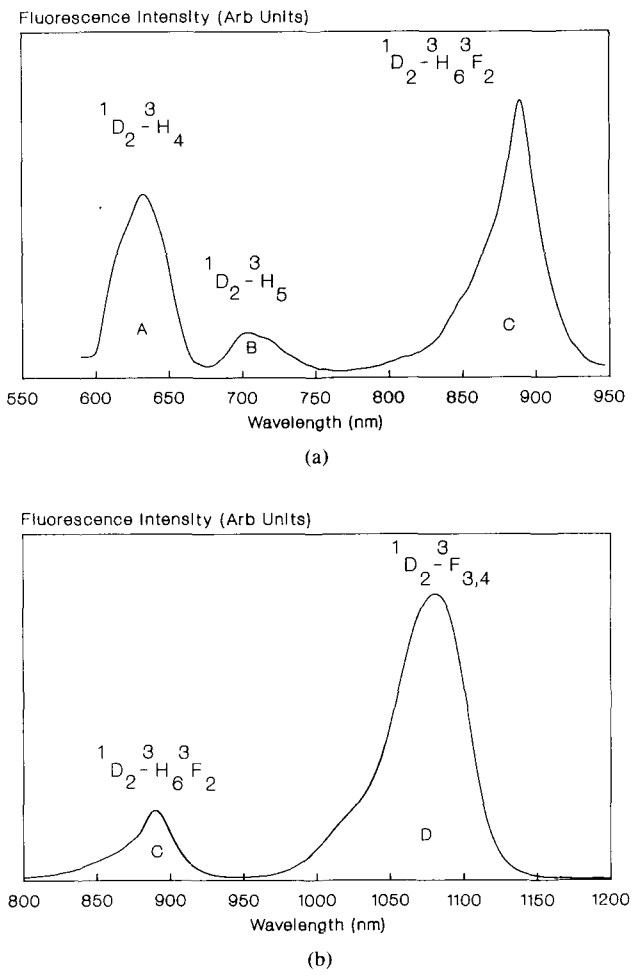

Fig. 6. (a) The fluorescence spectrum of Pr^{3+} in fused silica 550–950 nm, recorded in end-light, under excitation from an Rh6*G* dye laser at 590 nm. (b) The fluorescence spectrum of Pr^{3+} in fused silica 800–1200 nm, recorded in end-light, under excitation from an Rh6*G* dye laser at 590 nm.

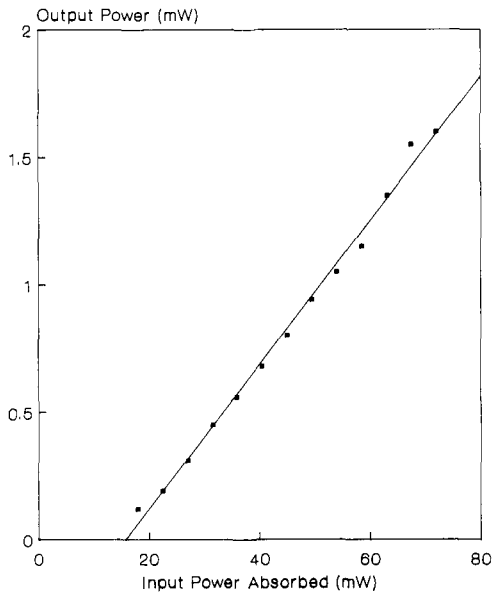

Fig. 7. The slope efficiency for laser oscillation at 1080 nm, under excitation at 590 nm.

high reflector. The slope efficiency with this cavity was 5 percent and the threshold was reached at $\simeq 5$ mW absorbed power. Knowing the threshold absorbed power, cavity loss and fluorescence bandwidth, one can use the equation [9],

$$\frac{P_{\text{th}}(\text{abs})}{h\nu_p} = (8\pi cn^2)\,\frac{A_{\text{eff}}\,\Delta\lambda}{\eta\beta\,\lambda_{em}^4}\,[\text{loss/pass}]$$

where η is the radiative quantum efficiency for the multiplet ($^1D_2$), and β is the branching ratio for the $^1D_2$–$^3F_{3,4}$ transition, to obtain a value for the product $\eta\beta$. Using the values $P_{\text{th}} = 2$ mW, loss/pass = 2 percent, $\Delta\lambda = 60$ nm at 1080 nm [from Fig. 2(b)], the product $\eta\beta$ is found to be $\simeq 2$ percent for the $^1D_2$–$^3F_{3,4}$ transition. Since the fluorescence bands span a large spectral range and could not all be recorded using the same monochromator grating and detector, correction of the spectrum for instrumental response has not be attempted. Given the uncertainty in spectral response it is thus impossible at this stage to give a value for the overall quantum efficiency of the $^1D_2$ multiplet.

To observe laser operation on the 888 nm transition a pair of mirrors with reflectivity >99.5 percent at 890 nm and a transmission of $\simeq 90$ percent at 1080 nm were used. The threshold for stimulated emission on this transition was then found to be $\simeq 10$ mW of absorbed power with overall cavity losses measured to be 2 percent. Again lasing occurred at several wavelengths over a range of $\simeq 10$ nm, this time centered at 888 nm. The relationship of output laser power and absorbed pump power was not investigated for this transition as no suitable output coupler was available. Using the above equation and the corresponding data on the 888 nm laser transition, the product $\eta\beta$ was determined to be $\simeq 0.7$ percent.

For both of these transitions the threshold for stimulated emission could be reduced by $\simeq 30$ percent by immersing all but a few centimeters of fiber in liquid nitrogen. A number of processes may contribute to this, the most significant is probably an improvement in the radiative efficiency at low temperatures.

The laser performance at 1080 nm was also investigated under laser pumping at 488 nm using an argon ion laser which excited the $^3P_0$ multiplet directly. Two mirrors with reflectivities of >99.5 and $\simeq 98$ percent at 1080 nm were used. The mirror at the input end of the fiber had a transmission of 90 percent for the pump radiation at 488 nm. The threshold observed with 488 nm pumping was $\simeq 30$ mW; some 15-fold higher than the threshold observed with 590 nm pumping. This suggests that the dominant decay routes from the $^3P_0$ multiplet do not populate the $^1D_2$ multiplet. Since the visible fluorescence excited by $^3P_0$ pumping appears to be weaker than with $^1D_2$ pumping it seems likely that nonradiative decays which bypass the $^1D_2$ multiplet are principally responsible for the poor performance. The mechanism cannot be identified at present but might involve a cross-relaxation process [10], particularly if the ions are clustered to any extent.

CONCLUSION

We have investigated the spontaneous and stimulated emission from praseodymium doped into a silica fiber and demonstrated laser action on two of these bands at 1080 and 888 nm, the latter for the first time. Either band in principle offers a tunable range of at least 50–60 nm [2]. However, our results show that the product of the quantum efficiency and branching ratio ($\eta\beta$) as indicated by the observed performance of the two lasing transitions is as low as 2 and 0.7 percent, on the $^1D_2$–$^3F_{3,4}$ and $^1D_2$–$^3F_2\,^3H_6$ transitions, respectively. This would tend to suggest that the probable quantum efficiency (η) for the $^1D_2$ multiplet is low. This incidentally illustrates the effectiveness of the fiber waveguide geometry in obtaining stimulated emission from laser transitions with low radiative quantum efficiencies.

In its present form a praseodymium-doped silica fiber does not look to be a promising laser system for application, particularly in view of its rapid nonradiative decay rates. However, in a glass host with lower phonon energies such as a fluoride glass, where nonradiative decay rates should be reduced, praseodymium may well prove to have a much improved performance.

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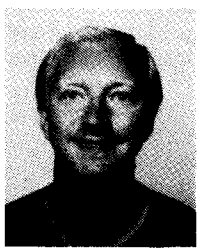

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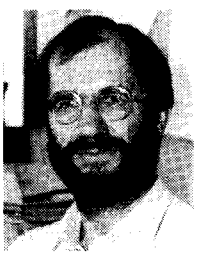

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