General procedure for the analysis of Er^{3+} cross sections

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We have applied the theory of McCumber to the $^4I_{13/2} \leftrightarrow {}^4I_{15/2}$ transition of Er^{3+}-doped glasses and confirmed its ability to provide both spectra and accurate values of cross sections. For an Al/P–silica fiber, the exact McCumber treatment is within 3% of the measured emission cross section, whereas an Einstein analysis is off by >50%. An approximate treatment that eliminates the McCumber theory's requirement of detailed electronic structure has been developed and found to usually provide more accurate values than an Einstein analysis.

The demonstration of the first single-mode Er^{3+} fiber amplifier at 1.5 μm (Ref. 1) generated great interest in such devices because they provide high gain and low noise at an important optical communications wavelength. As with all lasers and amplifiers, accurate cross sections are essential to the quantitative understanding of devices as well as the identification of improved active media. Despite the extensive work on Er^{3+} fiber amplifiers, however, uncertainty still surrounds the values of the cross sections. The accepted procedure for connecting absorption and stimulated-emission cross sections uses the relationship between the Einstein A and B coefficients generalized to account for a finite linewidth. Although this approach has been applied to Er^{3+}-doped glasses,[2,3] there is increasing evidence that it fails for the $^4I_{13/2} \leftrightarrow {}^4I_{15/2}$ transition.[4–6] We present an alternative analysis technique using the more general theory of McCumber,[7] which has been successful in the study of transition metal ions but seldom applied to rare earths. We show that this theory is, in fact, ideally suited to rare-earth ions and provides not only absolute cross sections but spectral information as well. Cross sections calculated according to the McCumber theory are in excellent agreement with experiments if the electronic structure of the ion is known. We find that even when an approximate technique is used to estimate the electronic parameters, it generally provides more accurate results than an Einstein analysis.

When it is not straightforward to measure both the absorption and stimulated-emission cross sections for a rare-earth-doped material, one is often calculated from the other using the Einstein A and B coefficients for a two-level system.[8] Generalized to the case in which the lower state (level 1) and upper state (level 2) are split into multiple components, the relationship becomes

$$g_1 \int \nu^2 \sigma_a(\nu) d\nu = g_2 \int \nu^2 \sigma_e(\nu) d\nu, \qquad (1)$$

where g_i is the degeneracy of level i, ν is the photon frequency, and σ_a and σ_e are the absorption and stimu-lated-emission cross sections, respectively. Equation (1), which is a more general form of the Ladenburg–Fuchtbauer relationship,[9] is valid for rare-earth ions only if one of two conditions is met. Either all components of the two levels must be equally populated, or all the transitions must have the same strength regardless of the components involved. Either of these conditions ensures that the observed oscillator strength is identical to that of the degenerate (unsplit) two-level case. Since the manifold widths of the $^4I_{15/2}$ and $^4I_{13/2}$ states of Er^{3+}-doped glasses are typically 300–400 cm^{-1},[10] the first condition is not satisfied at room temperature ($kT \approx 200$ cm^{-1}). Moreover low-temperature absorption and emission measurements indicate that for all glasses the transition strength is quite sensitive to the Stark levels involved.[10,11] Since neither of the above requirements is satisfied, it is not surprising that significant deviations from Eq. (1) have been reported.[4–6]

In contrast, the only assumption required by the theory of McCumber is that the time required to establish a thermal distribution within each manifold be short compared with the lifetime of that manifold. From consideration of detailed balance, the absorption and emission cross sections are then related by[7]

$$\sigma_e(\nu) = \sigma_a(\nu)\exp[(\epsilon - h\nu)/kT], \qquad (2)$$

where ϵ is the temperature-dependent excitation energy. The physical interpretation of ϵ is as the net free energy required to excite one Er^{3+} ion from the $^4I_{15/2}$ to the $^4I_{13/2}$ state at temperature T. Since the manifold widths exceed kT, the absorption and emission spectra are offset from each other, the absorption to higher frequency and the emission to lower frequency, as illustrated in Fig. 1. The spectra have been plotted as a function of wavelength to facilitate comparison with previously reported results. An examination of Eq. (2) reveals that the cross sections are equal only at the crossing frequency, $\nu_c \equiv \epsilon/h$. At frequencies higher than ν_c, the emission cross section is smaller than the absorption cross section and vice versa for $\nu < \nu_c$. Another important characteristic of Eq. (2) is that
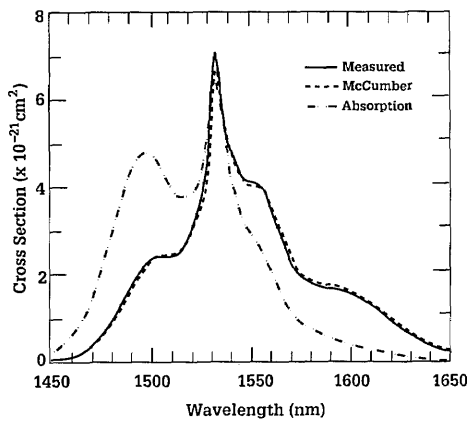
Fig. 1. Comparison of the shape of the measured stimulated-emission cross section with that calculated from the absorption cross section using the McCumber theory. The absorption spectrum used is also included. The glass is low-fluorine fluorophosphate composition L11.

even a relative measurement of either cross-section spectrum is sufficient to generate a relative spectrum of the other cross section, a feature clearly not available from the Einstein relation in Eq. (1).

To assess the McCumber theory's applicability to Er^{3+}-doped glasses, we compared its predictions with experimentally determined cross sections for four different glass compositions. This included examining both its ability to provide spectral information and the accuracy of the calculated values. $\sigma_a(\nu)$ was determined from absorption measurements on bulk samples with a known Er^{3+} concentration. A more involved procedure using measured radial distributions of the Er^{3+} ions and the optical mode was required to determine $\sigma_a(\nu)$ for the Al/P-doped silica fiber.[12] From corrected emission spectra and measured lifetimes for the $^4I_{13/2}$ state, the experimental stimulated-emission cross sections were found by using the expression[7]

$$\frac{1}{\tau} = \frac{8\pi n^2}{c^2} \int \nu^2 \sigma_e(\nu) d\nu, \tag{3}$$

where τ is the radiative lifetime and n is the refractive index.

We first examined the ability of Eq. (2) to produce the correct spectral shape. Measured absorption cross sections were transformed into relative emission cross-section spectra with Eq. (2) and converted into absolute cross sections by means of Eq. (3) and the measured lifetimes. Figure 1 shows that the shape of $\sigma_e(\nu)$ calculated in this way for a fluorophosphate glass agrees extremely well with that of the experimentally determined spectrum. The only significant discrepancy occurs at the peak, which is wider and lower for the calculated spectrum, a pattern that holds for the other three glasses investigated.

In addition to providing relative cross-section spectra, the McCumber theory can also provide absolute values if the parameter ϵ can be evaluated. For this purpose a useful alternative definition is[7]

$$\frac{N_1}{N_2} = \exp(\epsilon/kT), \tag{4}$$

where N_i is the equilibrium population of the ith level at temperature T in the absence of optical pumping. The low symmetry of the crystal field lifts all but the Kramers degeneracy of the Er^{3+} ion and splits the ground state into eight components and the excited state into seven components. If the positions of all the Stark components are known, ϵ is calculated according to

$$\frac{N_1}{N_2} = \frac{1 + \sum_{j=2}^{8} \exp(-E_{1j}/kT)}{\exp(-E_0/kT)\left[1 + \sum_{j=2}^{7} \exp(-E_{2j}/kT)\right]}. \tag{5}$$

In this expression E_0 is the separation between the lowest component of each manifold and E_{ij} is the difference in energy between the jth and the lowest component of level i. To test Eqs. (2), (4), and (5) we used a relative cross-section spectrum derived from the corrected emission spectrum of the Al/P–silica fiber. The measured $\sigma_e(\nu)$ was scaled by using Eq. (3) and the radiative lifetime, while that for the McCumber analysis was scaled using Eq. (5). The electronic structure used was that reported for a similar Al/Ge–silica fiber,[13] with the missing Stark components filled in by using data from a silicate glass.[10] The peak of the calculated $\sigma_e(\nu)$ is 5.9×10^{-21} cm^2, which is within 3% of the measured value indicated in Table 1. This confirms that McCumber theory can be applied to rare-earth ions by using the full generality expressed in Eq. (2). With the calculated value, the ratio of the absorption to the emission cross section at the peak of the latter is 1.1, in good agreement with the ratio of 1.2 ± 0.1 reported by Barnes et al. for a similar fiber.[6]

The electronic structure required for Eq. (5) is usually difficult to obtain. To eliminate this requirement we have developed a simple phenomenological procedure that yields a good estimate of ϵ for most of the glasses examined. We first assume a simplified electronic structure in which the Stark levels for a given manifold are equally spaced, $E_{ij} = (j - 1)E_i$. This reduces the number of parameters in Eq. (5) from 14 to 3: the component spacing for each manifold, E_i, and the separation between the two manifolds, E_0. In room-temperature measurements the peaks of the absorption and emission spectra are almost always within 5 cm^{-1} (1 nm) of each other. Low-temperature measurements[10,13] confirm that this wavelength corresponds to transitions between the lowest component

Table 1. Comparison of Procedures for Determining σ_e

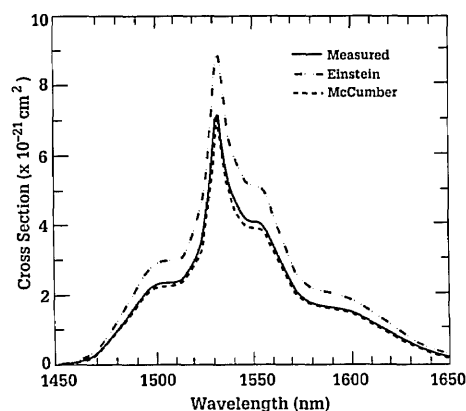| Glass	Peak σ_e (10^{-21} cm^2)		
	Measured	McCumber (approximate)	Einstein
Al/P–silica fiber	5.70	7.27	8.59
Silicate (L22)	7.27	8.46	7.72
Fluorophosphate (L11)	7.16	6.86	9.06
Fluorophosphate (L14)	5.79	5.71	7.62

Fig. 2.   Comparison of stimulated-emission cross sections scaled using the radiative lifetime (measured), the Einstein relation (Einstein), and the McCumber theory with an esti-mated ϵ (McCumber). The glass is fluorophosphate L11.

of each manifold. Accordingly, we take as E_0 the average of the absorption and emission peaks. If low-temperature measurements were available, the level-1 bandwidth would be the separation between the highest- and lowest-energy emission lines. Therefore we use for $7E_1$ the low-energy half-width of the room-temperature emission spectrum since the peak has already been identified with the transition between the lowest components of the two manifolds. In a similar way, the high-energy half-width of the absorption spectrum is adopted for the width of the excited-state manifold, $6E_2$. Other possible definitions in terms of half-bandwidths of emission and absorption spectra were tried and found to produce similar though less quantitative results. For the half-widths we use the separation between the peak and the energy at which the spectrum falls to 5% of the peak, although the results are not very sensitive to this criterion.

Figure 2 compares stimulated-emission cross sections determined from the corrected emission spectrum with three different scaling procedures. The measured curve was scaled by using the lifetime as above, the McCumber curve was scaled by using the above technique for estimating ϵ, and the Einstein curve was calculated using Eq. (1). The McCumber curve is in good agreement with the measured cross-section spectrum, whereas the Einstein curve considerably overestimates it. The results for the four glasses are summarized in Table 1, which compares the peak values of the emission cross section determined in the same ways as in Fig. 2. Only for the L22 silicate glass does the Einstein relation yield a more accurate value than the approximate technique for estimating ϵ; for the other glasses the Einstein treatment overestimates $\sigma_e(\nu)$ by at least 25%. As was reported previously,[5,6] the use of Eq. (1) leads to a particularly large error for silica fiber. In comparison, McCumber theory with an approximated ϵ does significantly better, while the McCumber theory with an accurate electronic structure yields values that agree within experimental error.

In summary, we have shown that the theory of McCumber is a powerful tool for analyzing cross sec-

tions of rare-earth-doped glasses. It provides not only absolute values but spectral information as well. Both these features have been tested and found to be in excellent agreement with experiment. A drawback of the McCumber theory is that it requires knowledge of the electronic structure of the dopant ion in order to provide absolute cross sections. We have demonstrated, however, that by the use of an extremely simple model of the spectra and electronic structure of Er^{3+}, values of ϵ can be obtained for the important $^4I_{13/2} \leftrightarrow$ $^4I_{15/2}$ transition that in most cases yield results significantly more accurate than those obtained from the Einstein relation. With this procedure for estimating ϵ, it is possible to generate either the absolute absorption or the emission cross-section spectrum from a measurement of the other. An application of the McCumber theory that exploits its ability to provide spectral information is to determine $\sigma_e(\nu)$ from measurements of $\sigma_a(\nu)$ and the radiative lifetime. The procedures outlined here can also be applied to excited-state transitions to give a relationship between quantities less experimentally accessible, such as excited-state absorption and emission spectra.

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