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Time-Resolved Measurement of Excited-State Populations in Atomic Systems

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Introduction The absorption of light is the most common method for the investigation of the energy levels and transfer processes of atomic and molecular systems. The measurement of the ground-state absorption (GSA) provides information on the energy level scheme. If the density of the atoms or molecules in the stable ground state is known then the cross sections of the GSA transitions can be calculated from a Lambert-Beer law (see (2) of the next Section). Once the cross section of a certain transition has been determined an unknown density of atoms can be ascertained by measuring the absorption of light on this transition.

The case is much more difficult if the populations of excited states and the cross sections of ESA are to be investigated because these states are unstable and decay, e.g. by the emission of light. In most cases it is impossible to obtain a defined distribution of the energy over the excited states and, therefore, the calculation of the ESA cross sections seems to fail. In this contribution we show that time-resolved ESA measurements, without the knowledge of the corresponding cross sections, provide enough information for the calculation of the excited-state populations. The ESA cross sections can then easily be calculated, and with the knowledge of the cross sections the distribution of the energy after any excitation can be derived.

Measuring method A pump- and probe-beam technique is used for the time-resolved measurement of excited-state absorption. The populations n_i of the excited states are achieved by pumping with a pulsed laser into one level. Subsequent relaxation populates lower lying excited states. n , $n_g(t)$, and $n_e(t)$ are the doping concentration, the time-depending population densities of the ground state and of all excited levels, respectively,

$$n = n_g(t) + n_e(t) = n_g(t) + \sum n_i(t). \quad (1)$$

The sum in (1) generally extends over all populated excited levels.

A pulsed probe beam of low intensity I_0 , such that the populations of the levels are unperturbed, passes through the sample of length d . The relevant processes induced by the probe beam are characterized by their cross sections σ_{GSA} (ground-state absorption) and σ_{ESA} (excited-state absorption). In the unpumped case all electrons are in the ground state and the transmitting probe beam intensity is

$$I_u(d) = I_0 \exp(-n\sigma_{\text{GSA}}d), \quad (2)$$

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whereas in the pumped case it is

$$I_p(d) = I_0 \exp(-d[n_g \sigma_{\text{GSA}} + \Sigma\{n_i \sigma_{\text{ESA}}(i)\}]) . \quad (3)$$

From (1) to (3) we calculate

$$\frac{1}{d} \ln(I_u/I_p) + n_c \sigma_{\text{GSA}} = \Sigma\{n_i \sigma_{\text{ESA}}(i)\} . \quad (4)$$

To obtain the populations n_i the terms on the left hand side of (4) must be determined. The cross sections σ_{ESA} do not have to be known. σ_{GSA} is obtained by measuring I_0 and I_u and using (2). The measurement of the transmitted probe beam without pump excitation or at adjustable delay times after pulsed laser excitation provides the quantities I_u and I_p , whilst n_c can be determined from the bleaching of the GSA. At wavelength regions where GSA but no ESA occurs we obtain from (4)

$$n_c = - \frac{1}{\sigma_{\text{GSA}} d} \ln(I_u/I_p) . \quad (5)$$

The populations n_i of the excited states can be derived from the following considerations: The sum of the absolute populations of all excited states at a certain delay time t_k after pulsed excitation is

$$\Sigma n_i(t_k) = n_c(t_k) . \quad (6)$$

The time-dependent measurements of ESA originating from m different levels provide the quantities

$$n_i(t_k) \sigma_{\text{ESA}}(i) ; \quad i = 1, \dots, m , \quad (7)$$

as demonstrated in (1) to (4). The comparison of the same ESA peak at different delay times t_1, t_k yields

$$\frac{n_i(t_1) \sigma_{\text{ESA}}(i)}{n_i(t_k) \sigma_{\text{ESA}}(i)} = \frac{n_i(t_1)}{n_i(t_k)} ; \quad i = 1, \dots, m , \quad k = 2, \dots, m . \quad (8)$$

So the temporal information replaces the information of the cross sections which are eliminated from the calculation. The measurement of ESA at m different delay times leads to m equations of form (6) and $m(m-1)$ independent equations of form (8). In total m^2 equations allow the determination of the m^2 quantities

$$n_i(t_k) ; \quad i, k = 1, \dots, m . \quad (9)$$

Finally, the cross sections $\sigma_{\text{ESA}}(i)$ are obtained from (4).

Results and discussion ESA spectra of Er^{3+} doped yttrium-aluminum-perovskite (YAlO_3) were measured at several delay times. According to the temporal behaviour of the ESA peaks the initial levels of the ESA in Er^{3+} were identified and the populations of these levels were calculated. Results for the metastable levels in Er^{3+} ($2.9 \times 10^{20} \text{ cm}^{-3}$): YAlO_3 are shown in Fig. 1. ESA from other levels was not detected because, due to the short lifetimes, the populations of these levels are very small. Detailed results on ESA cross sections and ESA effects in the laser system $\text{Er}^{3+} : \text{YAlO}_3$ will be published elsewhere /1/.

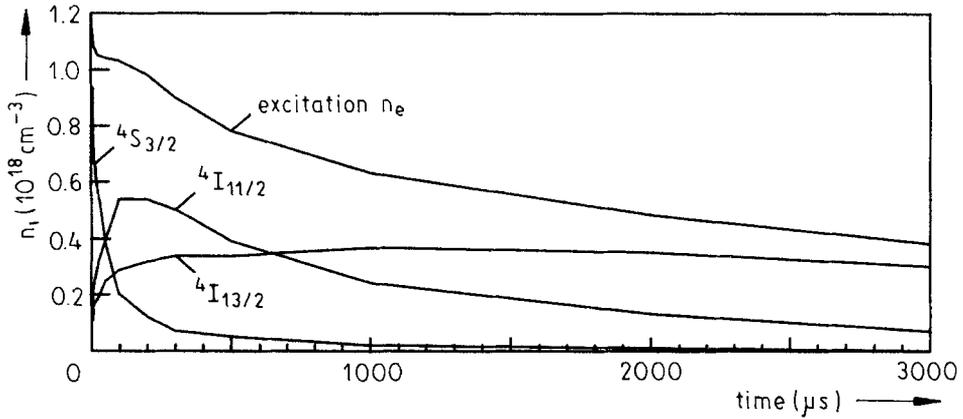


Fig. 1. Populations of the metastable excited states in $\text{Er}^{3+}:\text{YAIO}_3$ after pulsed excitation

Conclusions In conclusion, we have demonstrated that, with the information of time-resolved excited-state absorption spectra, the absolute populations of the excited states and the cross sections of the ESA transitions can be calculated.

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Reference

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