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Resonance radiation trapping effects in copper and manganese lasers

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Resonance radiation trapping effects in copper and manganese pure metals and their halides as laser materials have been investigated using Holstein's theory of resonance radiation trapping. Theoretical calculations have been performed for determining the laser-starting and optimum temperatures, and dissociation levels in the metal halides. The results are found to be in excellent agreement with the reported experimental values. A detailed discussion of the results has been presented.

INTRODUCTION

Laser action in copper has been observed by many investigators on the 5106- and 5782-Å transitions at temperatures of about 1800 K.¹⁻⁴ It has been reported that the $^2P_{3/2}$ - $^2D_{5/2}$ (5106 Å) transition required a tube temperature exceeding about 1475 K to sustain population inversion. A peak power of about 40 kW with an effective pulse width of 16 ns was obtained in a single-pulsed pure copper metal vapor laser at about 1800 K (Ref. 3). Laser action was also reported in pure manganese metal⁵ at 1120 K on the 5341-Å transition.

In addition, the use of metal halides to obtain pulsed laser emission from metal atoms using double-pulsed excitation has received considerable attention during the past 12 years due to their low operating temperatures. Laser action was reported at 485- and 875-K temperatures for copper chloride and manganese chloride, respectively, in double-pulsed excitations.⁶⁻⁸ In addition, metal oxides and organo-metallic compounds have also been reported as laser materials.^{9,10}

This approach to metal vapor lasers is attractive, as it could lead to the development of practical, efficient, and high-energy laser devices in the visible region of the spectrum. In these double-pulsed metal vapor lasers, the electrical dissociation of the metal compounds, the electron impact excitations of the metal atoms, and the radiation trapping upon the resonance lines are the principal inversion mechanisms. Weaver *et al.*¹¹ have reported the effective lifetimes of $^2P_{3/2}$ and $^2P_{1/2}$ energy levels of copper in CuI as well as the threshold ground-state densities required for radiation trapping upon 3248- and 3274-Å resonance lines. They have also estimated the reservoir temperature and the dissociation level at which the stimulated emission is observed. No such calculations are available in the literature for other metals and metal halides.

In this paper, we present the theoretical resonance radiation trapping threshold temperatures, and the temperatures at the beginning of the lifetime saturation in copper and manganese pure metals and their halides as laser materials. These temperatures are compared with the reported values of laser-starting and optimum temperatures. The present calculations are found to be in excellent agreement with the reported laser-starting and optimum temperatures in these

materials. The laser-optimum temperatures are found to occur at the beginning of the lifetime saturation of the upper-laser level. All these calculations have been carried out by using Holstein's theory of resonance radiation trapping.^{12,13} These calculations will guide future experimental studies in the most promising directions.

METHOD OF CALCULATION

The absorption coefficient (k_{ij}) upon the resonance radiative transitions for different temperatures is given by

$$k_{ij} = \frac{1}{8\pi} \sqrt{\frac{\ln 2}{\pi}} \frac{\lambda_{ij}^2 A_{ij} N_j g_j}{\Delta\nu_D g_i}, \quad (1)$$

where λ_{ij} is the wavelength of the resonance radiative transition, A_{ij} is the spontaneous atomic transition probability, N_j is the ground-state atomic population density, and g_j and g_i are the statistical weights of the ground-state and upper-laser levels. $\Delta\nu_D$ is the Doppler half-width at half-maximum (HWHM) of the radiative transition and is given by

$$\Delta\nu_D = 7.162 \times 10^{-7} \nu_0 \sqrt{T/M}, \quad (2)$$

where ν_0 is the central frequency of the transition, M is the atomic mass, and T the absolute temperature.

The atomic density in the ground state N_j is calculated using $N = P/KT$, where P is the molecular vapor pressure at temperature T , N is the molecular vapor density, and K is Boltzmann constant. The values of P have been computed by the least-squares fit for the standard equation¹⁴

$$\log P = A + B/T. \quad (3)$$

From the values of N , the ground-state atomic density of the metals N_j for different dissociation levels of the metal compounds can be computed.

Using Eq. (1), k_{ij} can be obtained for different dissociation levels at each temperature. The reabsorption of resonance radiation which will reduce the effective spontaneous emission rate to the ground state is given by

$$A_{ij}(\text{trapped}) = FA_{ij}(\text{untrapped}), \quad (4)$$

where F , the reduction factor, for a cylindrical volume of radius R is given by¹³

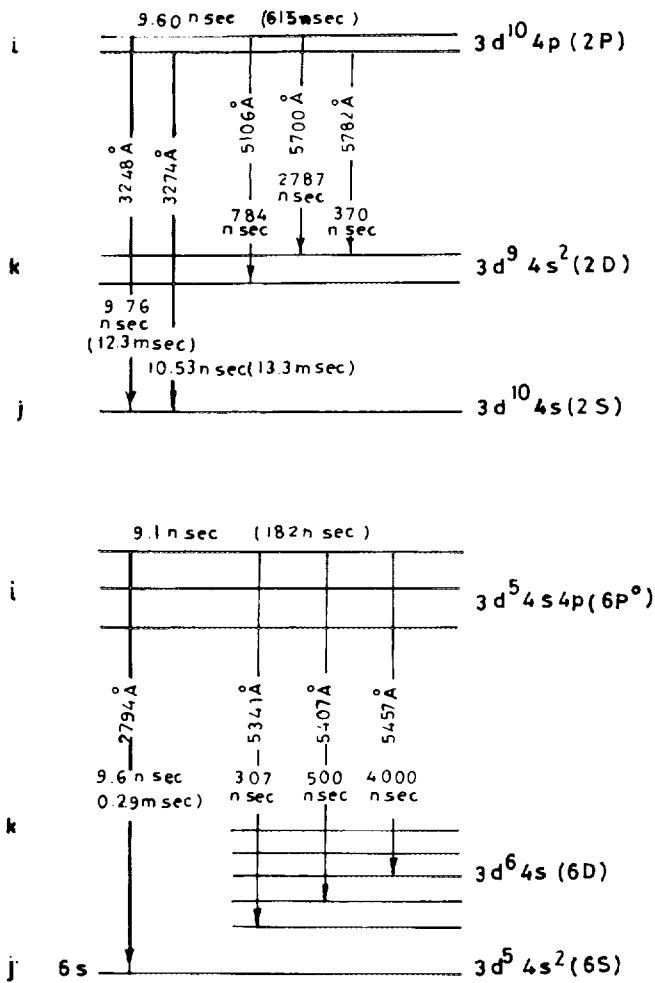


FIG. 1. Energy level diagrams of copper and manganese. Extended lifetimes and time constants are given in brackets.

$$F = \frac{1.6}{k_{ij} R [\pi \ln(k_{ij} R)]^{1/2}} \quad (5)$$

The resonance radiation trapping threshold temperature, threshold ground-state atomic densities, and threshold reduction factor can be calculated for the condition

$$(A_{ij})_{th} = A_{ik} + A_{ik'} + \dots \quad (6)$$

The corresponding threshold ground-state density represents the boundary between trapped and untrapped conditions. Above this value, resonance trapping effects would influence the upper-laser-level lifetimes. The upper-laser-level lifetime is then

$$\frac{1}{\tau} = \frac{1}{\tau_{ij}} + \left(\frac{1}{\tau_{ik}} + \frac{1}{\tau_{ik'}} + \dots \right), \quad (7)$$

where the energies (E) of i, k, k', \dots, j levels follow

$$E_i > E_k > E_{k'} \rightarrow E_j.$$

Using these equations, the upper-laser-level lifetimes can be calculated for different temperatures and dissociation levels.

RESULTS AND DISCUSSION

The energy-level diagrams of copper and manganese atoms are shown in Fig. 1. The resonance radiative transitions in copper (3248 and 3274 Å) and manganese (2794 Å) are indicated in bold lines. The laser transitions considered are 5106 and 5782 Å in copper and 5341 Å in manganese, which are shown by thin lines.

The vapor pressure data of the metals and metal compounds have been computed for Eq. (3) (Ref. 14) by using the least-squares fit method, from the data given in the literature.¹⁵ The vapor pressure data for metals have been taken, and upper-laser-level lifetimes have been computed with temperature variations, using Eqs. (1)–(7). All these calculations have been performed for a tube radius $R = 1$ cm. In

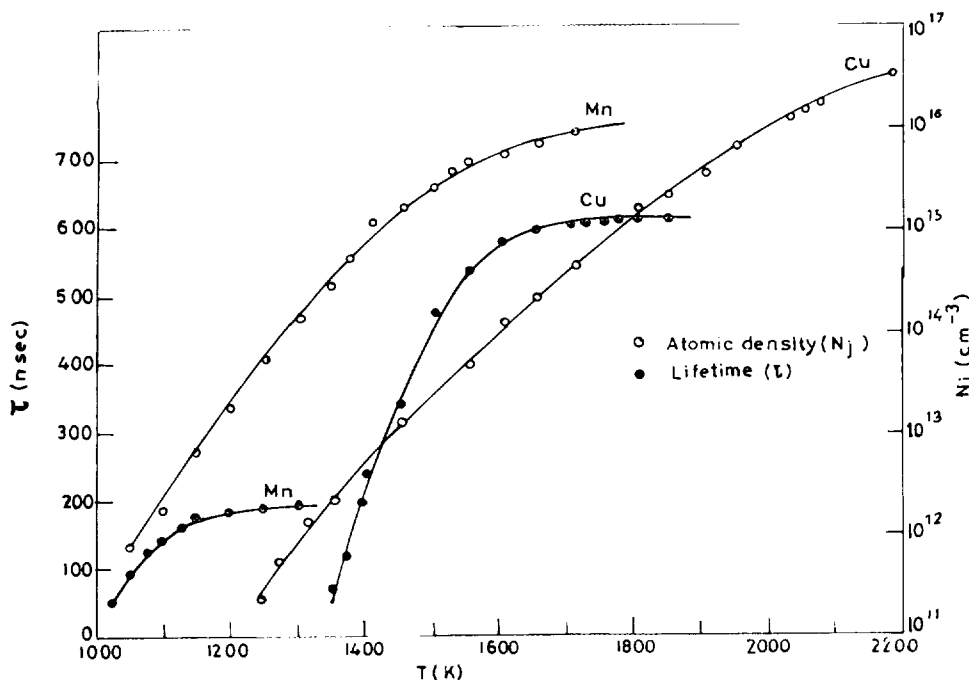


FIG. 2. Variation of lifetime and atomic density as a function of temperature.

TABLE I. Resonance radiation trapping threshold parameters.

System	F_{th} ($\times 10^{-2}$)	$(N_j)_{th}$ ($\times 10^{13} \text{ cm}^{-3}$)	τ_{th} (ns)	T_{th} in K		Optimum temperature (in K)		Dissociation level (in %)
				Reported	Present work	Reported	Present work	
(1) Cu ^a	1.58	1.5	308	1475 ^b	1425	...	1620	...
(2) CuCl ^a	1.58	1.5	308	590 ^c	585	673	665	10
(3) CuBr ^a	1.58	1.5	308	693 ^d	685	760	775	1
(4) CuI ^a	1.58	1.5	308	693 ^c	696	870	800	10
(5) Mn ^f	5.30	0.092	91	1120 ^g	1050	...	1175	...
(6) MnCl ₂ ^f	5.30	0.092	91	883 ^h	880	975	1050	0.01

^a Wavelength considered for copper 3248 Å.

^b Ref. 2.

^c Ref. 7.

^d Ref. 16.

^e Ref. 11.

^f Wavelength considered for manganese 2794 Å.

^g Ref. 5.

^h Ref. 8.

the case of copper, only the resonance radiation 3248 Å and corresponding laser wavelength 5106 Å have been taken into account for the calculations.

Figure 2 shows the variation of the upper-laser lifetime with temperature in the pure metals of copper and manganese. The upper-laser-level threshold lifetimes (τ_{th}) in these metals are determined using the condition for threshold trapping given in Eq. (6). The corresponding resonance radiation trapping temperature (T_{th}) at τ_{th} is found from Fig. 2 and tabulated in Table I.

The ground-state atomic density at trapping threshold ($(N_j)_{th}$) is found, corresponding to T_{th} from the N_j -temperature plots shown in Fig. 2 for these pure metals. These

threshold densities are taken to be the same for the metal atoms dissociated from their halides, as the required atomic density at the threshold is the same for pure metals and their halides.

The ground-state atomic densities N_j of these metals from the metal halides have been calculated from their molecular densities computed from the vapor pressure equation¹⁴ for different dissociation levels of 0.01, 0.1, 1.0, 10, and 100 at each temperature. As CuCl, CuBr, and CuI volatilize in the trimeric form, three copper atoms are available for each molecule vaporized. This situation has been taken into consideration for our calculations. The dependencies of the ground-state atomic density N_j upon the reduction fac-

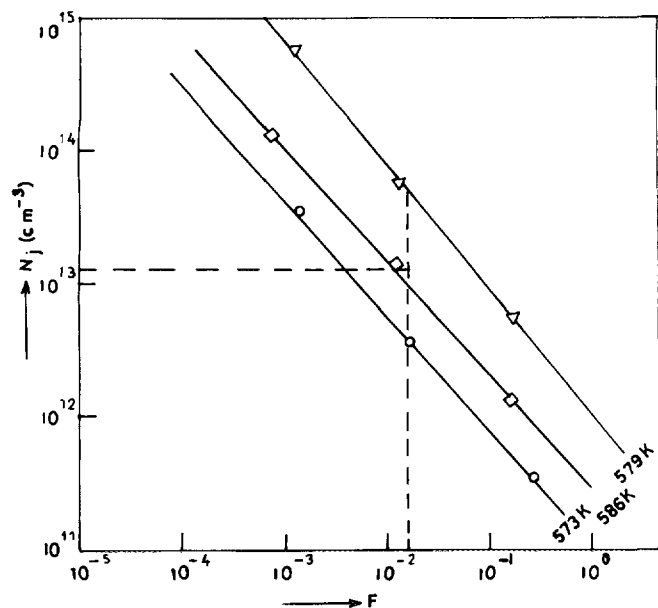


FIG. 3. The dependence of the ground-state atomic density N_j upon the reduction factor F in CuCl.

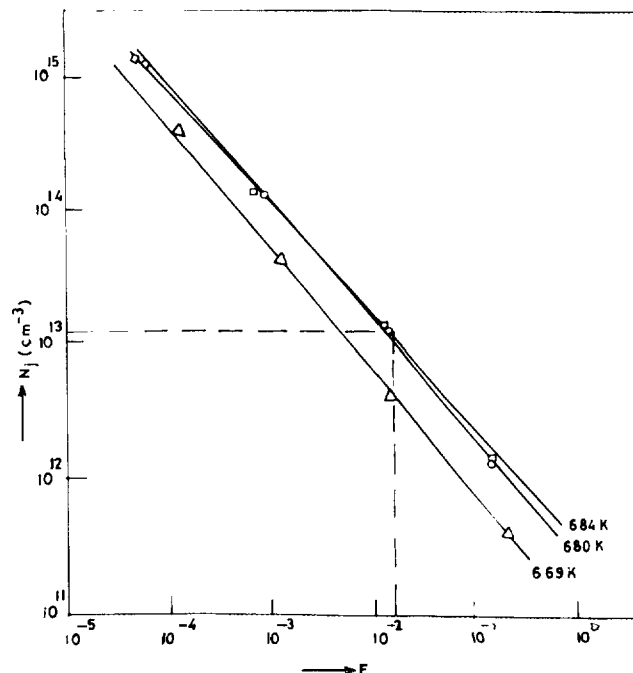


FIG. 4. The dependence of the ground-state atomic density N_j upon the reduction factor F in CuBr.

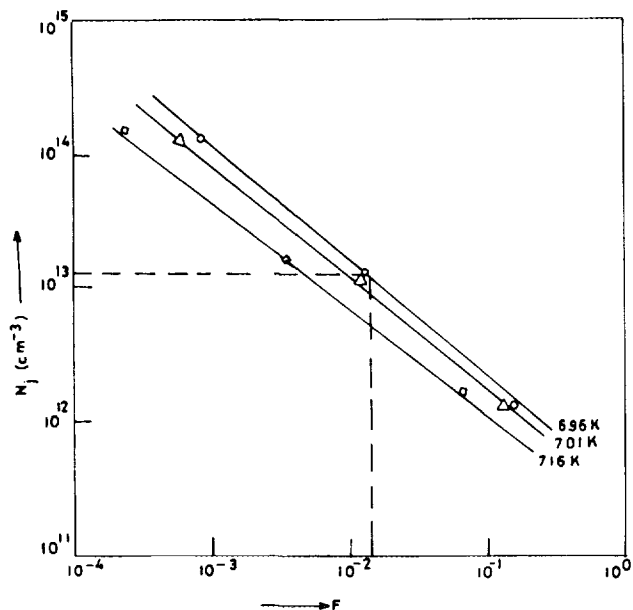


FIG. 5. Variation of the ground-state atomic density N_j with the reduction factor F in CuI.

tor F at different temperatures for CuCl, CuBr, CuI, and $MnCl_2$ have been drawn in Figs. 3–6. The value of the reduction factor at threshold (F_{th}) has been determined independently using

$$F_{th} = (A_{ik} + A_{ik'})/A_{ij} \text{ (ut) [from Eq. (4)] .}$$

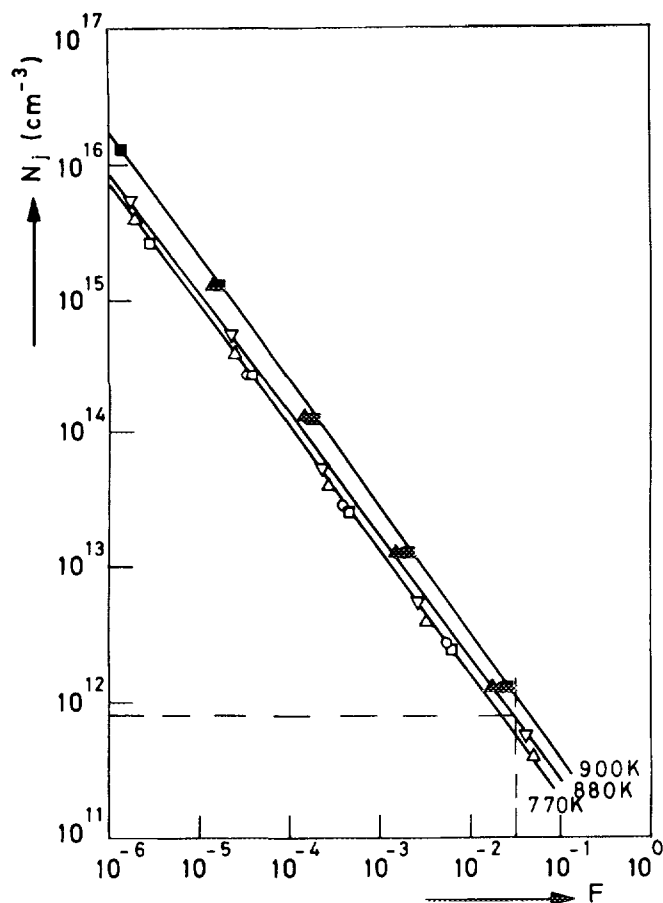


FIG. 6. Variation of the ground-state atomic density N_j with the reduction factor F in $MnCl_2$.

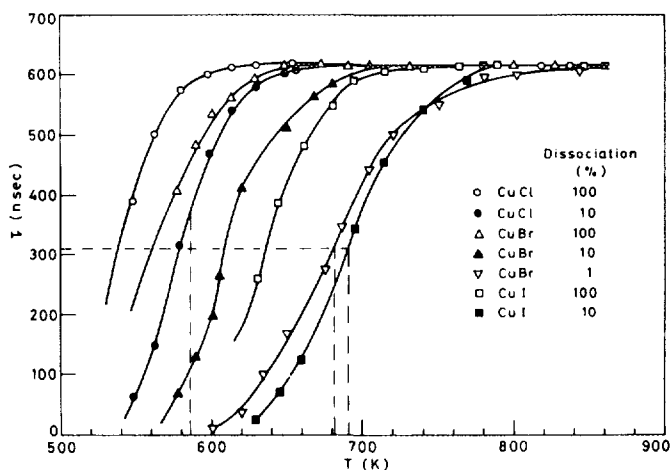


FIG. 7. The variation of the upper-laser lifetime (τ) with temperature for different dissociation levels in CuCl, CuBr, and CuI.

(N_j)_{th} and F_{th} are indicated with dotted lines in the figures. The meeting point of these two dotted lines is expected to be very close to the plot corresponding to one of the temperatures, and can be taken as the temperature of the trapping threshold of the resonance radiation. These values are shown in Table I.

The experimental laser-starting temperatures reported earlier for these compounds through double-pulsed excitations are given in column 5 of Table I for comparison. We have determined that the calculated values of T_{th} should correspond to the laser-starting temperatures, as the stimulated emission is expected to start at these temperatures. The calculated values are found to be in good agreement with the reported values within an accuracy of 5%.

Figures 7 and 8 show the variation of the upper-laser

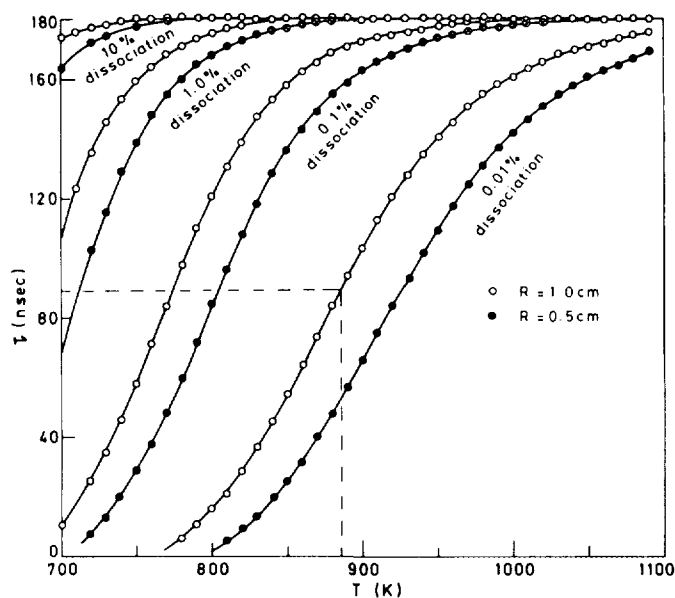


FIG. 8. The variation of the upper-laser lifetime (τ) with temperature for different dissociation levels in $MnCl_2$.

lifetime (τ) with temperature (T) for CuCl, CuBr, CuI, and MnCl₂ for different dissociation levels. The particular dissociation levels corresponding to τ_{th} and T_{th} have been determined from these plots, and found to be 10% in CuI, $\approx 10\%$ in CuCl, and 1% in CuBr. In MnCl₂ the dissociation level is found to be 0.01%. The dissociation level of 10% in CuI is consistent with that of the value given by Weaver *et al.*¹¹ It is quite interesting to find from these figures, corresponding to these particular dissociation levels, that the temperatures at the beginning of the lifetime saturation are approximately the same as the optimum temperatures reported earlier. A possible explanation is yet to be given to confirm this observation. Experimental investigations by double-pulsed laser excitations using these metal halides are in progress, systematically, to establish the validity of these theoretical calculations.

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