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Prepared for the
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~~The University of Arizona~~
Arizona Research Labs, *TUCSON*

Professor Marlan O. Scully, Principal Investigator

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ABSTRACT

Several aspects of the generation of x-rays in a crystal were investigated in order to explain some of the experimental results observed by Das Gupta. Sequential excitation of planes of two-level atoms in a crystal was shown to produce stimulated line radiation whose intensity varied exponentially with the incident electron beam current. This result agreed qualitatively with the experimental data. Swept excitation was also used to explain the increase in the amount of line radiation produced at a given electron beam current when the electron's energy was changed from 8.95 KeV to 16.997 KeV. The difference between the lifetime of an excited two-level atom in free space and the lifetime of the same atom in a lattice of nonresonant two-level atoms was also investigated. The resulting difference was attributed to Bragg scattering of the emitted radiation in the crystal and the bulk properties of the material.

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The purpose of this research has been to explain the experimental results obtained by Das Gupta.¹ These results include a nonlinear increase in the amount of K_{α} radiation emitted from a copper target and a decrease in the K_{α} linewidth when the intensity of the incident electron or x-ray beam is increased.

Our research has been oriented towards two major areas of investigation. In the first, sequential excitation of the atoms in a crystal has been invoked as a possible means of explaining the nonlinear increase in the K_{α} intensity. The results of calculations for a crystal that consists of a linear array of atoms shows that the intensity of the emitted radiation increases exponentially as the intensity of the pump beam is increased. This result is in qualitative agreement with the experimental data. Quantitative comparisons are meaningless until further work is completed. In particular, the effects of the three-dimensional crystal lattice on the direction and linewidth of the emitted radiation must be taken into account. In order to study the linewidth, the second area of research was directed towards an investigation of the properties of the radiation spontaneously emitted by an excited atom in a crystal. The preliminary results of this calculation show that the lifetime of an excited two-level atom in a crystal of nonresonant two-level atoms is different from the lifetime of the same excited atom in free space. This work may have also provided the first steps towards understanding the linewidth narrowing observed by Das Gupta.¹ Further work on the stimulated emission linewidth of an excited atom in a crystal is necessary before the relevance of this approach can be determined.

The nonlinear rise in the amount of K_{α_1} and K_{α_2} radiation emitted from a copper target has been seen using both electrons¹ and x-rays² to excite the copper atom. Because beams of both charged and neutral particles produce the same nonlinear response, the mechanism responsible for this increase is to first order independent of the method of excitation. In particular, this means that those effects that rely exclusively on a charged excitation beam need not be considered at first. The elimination of charged particle effects such as stimulated quasi-Cherenkov radiation and channeling greatly simplifies the problem.

K_{α_1} radiation is produced in a two step process. In the first step a $1s$ electron is removed from the atom by the pump. A $2p_{3/2}$ electron then makes a radiative transition to a $1s$ state producing a K_{α_1} photon. The dominant competing process is Auger recombination where the energy released by the $2p_{3/2} \rightarrow 1s$ transition removes a higher level electron from the atom thus leaving the atom doubly ionized. The Auger recombination time τ_a is $\sim 10^{-15}$ sec. This means that for an initially excited medium only a volume of dimensions $c\tau_a \sim 3 \times 10^{-5}$ cm could participate in stimulated scattering. This is much smaller than the characteristic dimension of a copper crystal $L \sim 10^{-3}$ cm reported by Das Gupta.² The limit imposed by the Auger recombination can be removed if the pumping is considered as swept gain excitation. This is the model of the excitation mechanism that we have chosen for the Das Gupta experiment. The experiment can then be modeled in terms of a two level system.

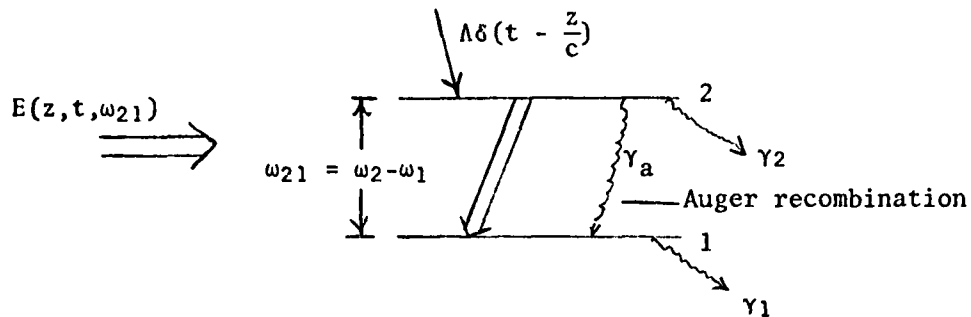


Figure 1

where 2 is the state with an electron missing in the $1s$ shell, 1 is the state with a filled $1s$ shell; γ_a is the Auger recombination rate; γ_2 , γ_1 are the decay rates of the two states; and Λ is the rate at which state 2 is populated.

The density matrix equation of motion along with Maxwell's equations form a closed set of equations that describe this two level system. These equations can be solved in the weak field limit (see Appendix A). The resulting expression for the intensity of the K_{α} radiation predicts an exponential increase as a function of Λ . This is at least in qualitative agreement with Das Gupta's results.¹ (If Das Gupta's data on the variation of the K_{α_1} intensity as a function of pump intensity is plotted on semilog graph paper then the resulting points very nearly lie on a straight line. This is to be contrasted with Das Gupta's plot). A quantitative comparison of the predictions of the model and the experimental data has not yet been made.

Das Gupta has recently told us that if the copper sample is excited with 16.997 KeV x-rays instead of 8.95 KeV x-rays then the nonlinear response of the $Cu K_{\alpha_1}$ line intensity to the pump beam intensity is even more pronounced. The two energies 8.95 KeV and 16.997 KeV for the x-ray pump beam corresponds to $E_{ion} = 8.95$ KeV which is the ionization energy for a $1s$ electron and $E = 16.997$ KeV = $E_{ion} + E_{K_{\alpha_1}}$ where $E_{K_{\alpha_1}} = 8.047$ KeV is

the energy of a Cu K_{α_1} photon. Because the pump beam is incoherent, the experiment with $E = 16.997$ KeV can be described in terms of two decoupled two-level systems as follows:

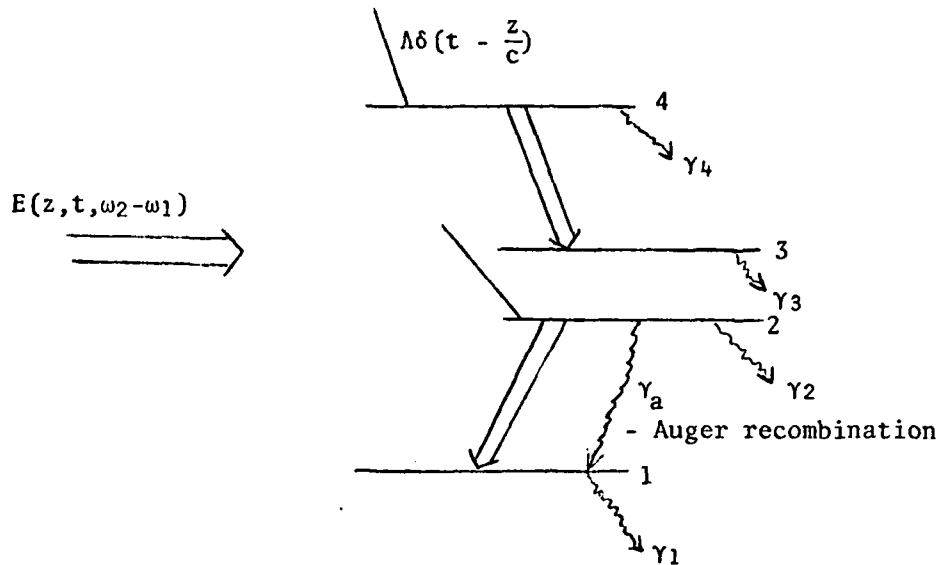


Figure 2

where $\hbar(\omega_4 - \omega_1) = 16.997$ KeV, $\hbar(\omega_2 - \omega_1) = \hbar(\omega_4 - \omega_3) = 8.047$ KeV and $\hbar(\omega_3 - \omega_1) = 8.950$ KeV. The two level system comprising states 1 and 2 is the same as described in Fig. 1. The remaining two level system is composed of states 4 and 3. State 4 is an energy state in the continuum 16.997 KeV above the 1s state of a copper atom. It is pumped at the same rate as state 2 because in the experiment the removal of a 1s electron by a 16.997 KeV photon not only creates an atom in state 2 but also leaves the ejected electron with a kinetic energy of 8.047 KeV in state 4. State 3 is an energy state at the ionization threshold. The density matrix equation for the two two-level systems plus Maxwell's equations again form a complete set. The resulting expression for the intensity of the copper K_{α_1} radiation again predicts an exponential increase as a function of the pumping rate (see Appendix B) in the weak field limit. For the case where there is no broadening mechanism

present, the difference between the gain coefficient when the energy of the pump beam is 16.997 KeV and the gain coefficient when the energy of the pump beam is 8.950 KeV is given by

$$\alpha(16.997 \text{ KeV}) - \alpha(8.950 \text{ KeV}) = \frac{\omega_{21}\mu_{12}^2 AN}{c\epsilon_0 h} \theta(\xi)f(\xi) ,$$

where μ_{12} is the dipole moment, N is the number of atoms per unit volume, $\xi = t - z/c$, θ is the unit step function and $f(\xi) = (e^{-\gamma_3 \xi} - e^{-\gamma_4 \xi})/(\gamma_2 - \gamma_1)$. The predictions of the above expression have not yet been compared with the experiment because Das Gupta has not yet completed the experiment.

In order to make a quantitative comparison between our two models and the results of the experiment it is necessary to understand what effect the crystal has on an excited atom. P. S. Lee and Y. C. Lee³ have already shown that if an excited two-level atom is placed in a crystal consisting of identical resonant, two-level atoms then the spontaneous decay rate of the excited state is vastly different from its free-space value. For the case of the Das Gupta experiments, the K_α radiation emitted by an initially excited atom is not resonant with the unexcited copper atoms in this crystal. A model relevant to the conditions of this experiment consists of a single excited two-level atom in a crystalline array of nonresonant two-level atoms. Approximate solutions for the probability amplitudes of the various states of the system have been found with the aid of the multiple time scale perturbation theory of Lindstedt and Poincare.⁵ With this method the probability amplitudes have been solved to fourth order in the coupling constants (see Appendix C for details). Of particular interest is the complex frequency of the amplitude for the initially excited atom. The expression for this frequency consists of five terms. Three of these terms

correspond to the complex frequency that would exist if the atom were in free space. The remaining two terms take into account the fact that the atom is in a medium. One of these terms is independent of the structure of the material whereas the other takes into account the lattice structure. It is this latter term which is of interest. It takes into account the Bragg scattering of the emitted radiation by the crystal. A rough estimate of the imaginary part of the complex frequency indicates it is less than 1% of the Weisskopf-Wigner linewidth and is therefore not important in most cases. The effect of the crystal on the stimulated linewidth has yet to be determined.

A report of our work on spontaneous emission of an excited atom in a crystalline array of nonresonant identical atoms was recently presented at the International Quantum Electronics Conference in June, 1980.

Our present results explain qualitatively some of the results of Das Gupta's experiments. Quantitative comparisons between the prediction of this model and the experiments cannot be made until the effects of the crystal are built into the swept gain model. In particular, the Bragg scattering of emitted x-ray radiation must be taken into account in order to make an accurate prediction of the K_{α} radiation measured in the experiment. This objection does not hold for the prediction of the increase in the gain coefficient resulting from increasing the energy of the pump beam from 8.95 KeV to 16.995 KeV. A quantitative comparison of our prediction and the results of the experiment await the completion of the experiment.

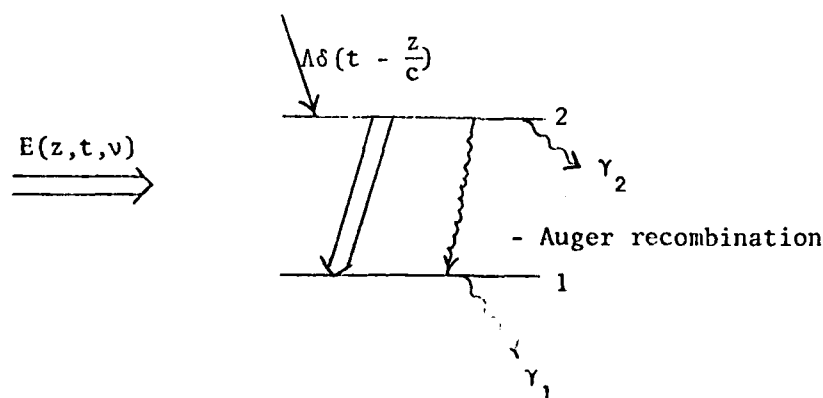
APPENDIX A

In the experiment an electron or an x-ray photon ionizes a copper atom by removing an electron from the 1s shell.¹ The vacancy in the 1s shell is removed either by a radiative transition of an electron from a higher level (2p electron in the case of K_{α} radiation) or by Auger recombinations. The Auger recombination is not a radiative transition. The energy released by a 2p electron going to the 1s vacancy is used to remove a higher lying electron from the atom thus leaving it doubly ionized. Since the electrons involved in these two processes are either in the 1s or 2p shell of the copper atoms, the atom can to a good approximation be modeled as a two-level atom.

The characteristic life time of the excited atom before it decays by Auger recombination is $\tau_a \approx 10^{-15}$ sec. This means that stimulated emission for a group of excited atoms will only occur for those atoms that are within $c\tau_a \sim 10^{-5}$ cm of each other. This is much smaller than the characteristic size 10^{-3} cm of a crystal in the sample used in this experiment.

One method of increasing the distance over which stimulated emission can occur is to sequentially excite the atoms along some axis. This is what is referred to as swept gain amplification.⁴

The various processes that occur in the two-level atom can then be represented by the diagram:



where state 2 is the excited state of the atom (an electron is removed from the inner shell), state 1 is the final state of the atom (1s shell is filled), γ_a is the Auger decay rate, γ_1 and γ_2 are the decay rate for the lower and upper states, $E(z,t)$ is the incident electromagnetic field of frequency ν and Λ is the rate at which the atoms are excited. Because these atoms are in a crystal Λ can be a function of z .

This system is described by the equations of motion for the density matrix elements

$$\dot{\rho}_{22} = \Lambda(z)\delta(t - \frac{z}{c}) - (\gamma_a + \gamma_2) \rho_{22} - \frac{i}{\hbar} (V_{21}\rho_{12} - V_{12}\rho_{21}), \quad (\text{A.1})$$

$$\dot{\rho}_{11} = \gamma_a \rho_{22} - \gamma_1 \rho_{11} - \frac{i}{\hbar} (V_{12}\rho_{21} - V_{21}\rho_{12}), \quad (\text{A.2})$$

$$\dot{\rho}_{21} = -i(\omega_2 + \Gamma)\rho_{21} + \frac{i}{\hbar} (\rho_{22} - \rho_{11})V_{21}, \quad (\text{A.3})$$

and the wave equation

$$\frac{\partial^2}{\partial z^2} E(z,t) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} E(z,t) = \mu_0 \frac{\partial^2 P}{\partial t^2}, \quad (\text{A.4})$$

$$P(z,t) = N\mu_{21}(\rho_{21} + \rho_{12}), \quad (\text{A.5})$$

where $V_{21} = -\mu_{21}E(z,t)$, $\mu_{21} = e\langle 2|x|1\rangle$, $\Gamma = (\gamma_1 + \gamma_a + \gamma_2)/2$, N is the number of atoms and $\hbar\omega_{21}$ is the difference in energy between states 2 and 1.

The electromagnetic field and polarization field can be rewritten in terms of a rapidly oscillating part and a slowly varying part as

$$E(z,t) = \mathcal{E}(z,t) \cos(kz - \nu t), \quad (\text{A.6})$$

$$P(z,t) = S(z,t) \sin(kz - vt), \quad (A.7)$$

with the conditions

$$k\mathcal{E} \gg \frac{\partial \mathcal{E}}{\partial z}, \quad v \gg \frac{\partial \mathcal{E}}{\partial t}, \quad ks \gg \frac{\partial S}{\partial z}, \quad vS \gg \frac{\partial S}{\partial t}. \quad (A.8)$$

We shall assume exact resonance $\omega_L = v$ from now on. The substitution of Eqs. (A.6) - (A.8) into Eq. (A.4) yields

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t} = \frac{v}{2c\epsilon_0} S, \quad (A.9)$$

where with aid of Eqs. (A.1) - (A.3) and (A.5) S is given by

$$S(z,t) = \frac{\mu^2 N}{h} \int_0^t dt (\rho_{22} - \rho_{11}) \mathcal{E}(z,t') e^{-\Gamma(t-t')}, \quad (A.10)$$

If $\mathcal{E}(z,t)$ is small (that is, we are interested in the linear gain region) then ρ_{11} and ρ_{22} are approximately given by

$$\dot{\rho}_{22} \approx \Lambda(z) \delta(t - \frac{z}{c}) - (\gamma_a + \gamma_2) \rho_{22},$$

$$\dot{\rho}_{11} \approx \gamma_a \rho_{22} - \gamma_1 \rho_{11},$$

so that

$$\rho_{22} \approx \Lambda(z) e^{-(\gamma_a + \gamma_2)\xi} \theta(\xi), \quad (A.11)$$

$$\rho_{11} \approx \Lambda(z) \gamma_a \begin{pmatrix} -(\gamma_a + \gamma_2)\xi & -\gamma_1 \xi \\ e & -e \\ \gamma_1 - (\gamma_a + \gamma_2) & \end{pmatrix} \theta(\xi), \quad (A.12)$$

where $\xi = t - \frac{z}{c}$ and $\theta(\xi) = 1$ when $\xi > 0$ and $= 0$ when $\xi < 0$. The combination of Eqs. (A.9) - (A.11) and (A.12) then give an equation for the amplitude of the electromagnetic field

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t} = 2r \int_0^t dt' \theta(\xi') \mathcal{E}(z, t') e^{-\Gamma(t-t')} \times e^{-(\gamma_a + \gamma_2)\xi'} + \frac{\gamma_a \left[e^{-\gamma_1 \xi'} - e^{-(\gamma_a + \gamma_2)\xi'} \right]}{\gamma_1 - (\gamma_a + \gamma_2)}, \quad (\text{A.13})$$

where $r = \nu \mu_{21}^2 N A(z) / c \epsilon_0 h$. This equation can be rewritten in terms of the amplitudes squared

$$\begin{aligned} \left(\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) \mathcal{E}^2(z, t) &= r(z) \mathcal{E}(z, t) \int_0^t dt' \left[e^{-(\gamma_a + \gamma_2)\xi'} + \frac{\gamma_a \left[e^{-\gamma_1 \xi'} - e^{-(\gamma_a + \gamma_2)\xi'} \right]}{\gamma_1 - (\gamma_a + \gamma_2)} \right] \theta(\xi') \mathcal{E}(z, t') e^{-\Gamma(t-t')} \\ &= r(z) \mathcal{E}(z, t) \theta(\xi) \left\{ e^{-(\gamma_a + \gamma_2)\xi} \left(1 + \frac{\gamma_a}{\gamma_2 + \gamma_a - \gamma_1} \right) \int_0^t dt' e^{\frac{(\gamma_a + \gamma_2 - \gamma_1)\tau}{2}} \right. \\ &\quad \left. \times \mathcal{E}(z, t - \tau) + \frac{\gamma_a e^{-\gamma_1 \xi}}{\gamma_1 - (\gamma_a + \gamma_2)} \int_0^\xi dt' e^{\frac{\gamma_1 - (\gamma_a + \gamma_2)\tau}{2}} \mathcal{E}(z, t - \tau) \right\}. \end{aligned} \quad (\text{A.14})$$

Because γ_1^{-1} and γ_a^{-1} are small in comparison to the time for $\mathcal{E}(z, t)$ to change significantly (the medium is excited by an essential continuous beam) then $\mathcal{E}(z, t - \tau) \approx \mathcal{E}(z, t)$ so that Eq. (14) becomes

$$\begin{aligned} \left(\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) \mathcal{E}^2(z, t) &= r(z) F(\xi) \mathcal{E}^2(z, t) \\ F(\xi) &= \theta(\xi) \left[\left(1 + \frac{\gamma_a}{\gamma_a + \gamma_2 - \gamma_1} \right) e^{(\gamma_a + \gamma_2 - \gamma_1)\tau/2} \right. \end{aligned}$$

$$+ \frac{\gamma_a}{\gamma_1 - (\gamma_a + \gamma_2)} \int_0^\xi d\tau e^{\frac{\gamma_1 - (\gamma_a + \gamma_2)}{2} \tau} \quad (A.15)$$

If we change variables from (z, t) to $(\xi = (t - \frac{z}{c}), z' = z)$ then

$$\frac{\partial}{\partial z} = \frac{\partial}{\partial \xi} \frac{\partial \xi}{\partial z} + \frac{\partial}{\partial z'} \frac{\partial z'}{\partial z} = \frac{\partial}{\partial z'} - \frac{1}{c} \frac{\partial}{\partial \xi} ,$$

$$\frac{\partial}{\partial t} = \frac{\partial}{\partial \xi} \frac{\partial \xi}{\partial t} = \frac{\partial}{\partial \xi} ,$$

so that

$$\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \rightarrow \frac{\partial}{\partial z'} ,$$

and

$$\frac{\partial}{\partial z'} \mathcal{E}^2(z', \xi) = r(z') F(\xi) \mathcal{E}^2(z', \xi) ,$$

which has a solution

$$\mathcal{E}^2(z', \xi) = \mathcal{E}^2(0, \xi) \exp\left\{ \int_0^{z'} r(x) dx F(\xi) \right\} .$$

(A.17)

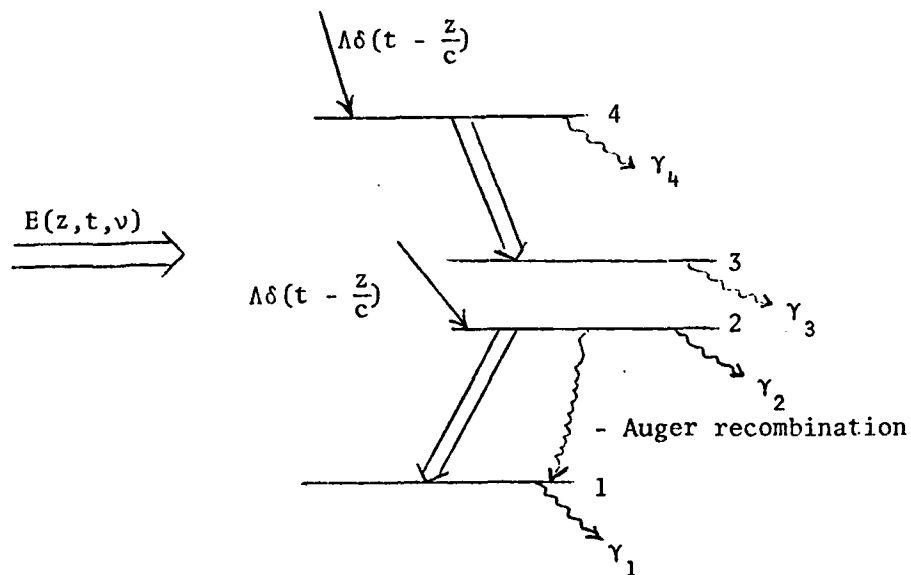
If $\Lambda(z) = \Lambda f(z)$ then we see that Eq. (A.17) predicts that the intensity of the K_{α_1} radiation will increase exponentially as the pumping rate Λ is increased. This result is in qualitative agreement with the results of Das Gupta's experiment.¹

It is important to note that Das Gupta has fitted his data to a polynomial in the incident beam intensity.¹ An equally good fit can be obtained by fitting the data to an exponential in the incident beam intensity. This is particularly true for the results of the K_{α_1} data.

Although this model qualitatively agrees with the experiment, a quantitative comparison is difficult and probably not worth while at the present stage. This model is only one-dimensional and therefore does not take into account the diffraction of the emitted radiation that will occur in the crystal. This is a rather serious shortcoming of the present model and is being rectified.

APPENDIX B

Das Gupta has recently begun an experiment where the copper sample is excited with 16.997 KeV x-rays instead of 8.95 KeV x-rays. The preliminary results of this experiment show that the nonlinear response of the Cu K_{α_1} intensity to the pump beam intensity is even greater. The quantity 8.95 KeV is the energy E_i necessary to ionize the copper atom by removing one electron from the 1s shell. The 16.997 KeV energy is the sum of E_i and $E_K = 8.047$ KeV which is the energy of a Cu K_{α_1} photon. Because the pump beam is incoherent, the experiment with $E = 16.997$ KeV can be described in terms of two decoupled two-level systems with swept excitation as follows:



where $E_4 - E_1 = 16.997$ KeV, $\hbar(\omega_2 - \omega_1) = \hbar(\omega_4 - \omega_3) = 8.047$ KeV and $\omega_3 - \omega_1 = 8.950$ KeV. The two level system consisting of states 2 and 1 is the same as was considered in Appendix A. States 4 and 3 are in the continuum. Although these two states should be represented by a band of states, we will consider only a single state in each region.

This system of two two-level atoms is described by the equations of motion for the density matrix elements and the wave equation

$$\dot{\rho}_{22} = \Lambda(z) \left(t - \frac{z}{c} \right) - (\gamma_a + \gamma_2) \rho_{22} - \frac{i}{\hbar} (V_{21} \rho_{12} - V_{12} \rho_{21}), \quad (\text{B.1})$$

$$\dot{\rho}_{11} = \gamma_a \rho_{22} - \gamma_1 \rho_{11} - \frac{i}{\hbar} (V_{12} \rho_{21} - V_{21} \rho_{12}), \quad (\text{B.2})$$

$$\dot{\rho}_{21} = -i \left(\omega_2 + \frac{\gamma_1 + \gamma_2 + \gamma_a}{2} \right) \rho_{21} + \frac{i}{\hbar} (\rho_{22} - \rho_{11}) V_{21}, \quad (\text{B.3})$$

$$\dot{\rho}_{44} = \Lambda(z) \delta \left(t - \frac{z}{c} \right) - \gamma_4 \rho_{44} - \frac{i}{\hbar} (V_{43} \rho_{34} - V_{34} \rho_{43}), \quad (\text{B.4})$$

$$\dot{\rho}_{33} = -\gamma_3 \rho_{33} - \frac{i}{\hbar} (V_{34} \rho_{43} - V_{43} \rho_{34}), \quad (\text{B.5})$$

$$\dot{\rho}_{43} = -i (\omega_2 + \gamma_3) \rho_{43} + \frac{i}{\hbar} (\rho_{44} - \rho_{33}) V_{43}, \quad (\text{B.6})$$

$$\rho_{12} = \rho_{21}^*, \quad \rho_{34} = \rho_{43}^*, \quad (\text{B.7})$$

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E(z, t) = \mu_0 \frac{\partial^2}{\partial t^2} P(z, t), \quad (\text{B.8})$$

$$P(z, t) = N [\mu_{21} (\rho_{21} + \rho_{12}) + \mu_{43} (\rho_{43} + \rho_{34})], \quad (\text{B.9})$$

where $V_{ij} = -\mu_{ij}E(z,t)$, $\mu_{ij} = \langle i|x|j\rangle$, $\gamma_{ij} = (\gamma_i + \gamma_j)/2$, $E(z,t,\omega_2)$ is an external electromagnetic field and $\hbar\omega_2 = 8.047$ KeV. We have chosen $\omega_1 = 0$.

The electric field and polarization field can again be written in terms of a rapidly varying part and slowly varying part as was done in Appendix A. The equation for the slowly varying part of the fields is then

$$\left(\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t}\right) \mathcal{E}(z,t) = \frac{v}{2c\epsilon_0} S, \quad (\text{B.10})$$

where S is given by

$$S = \frac{N}{\hbar} \left[\mu_{21}^2 \int_0^t dt' (\rho_{22} - \rho_{11}) \mathcal{E}(z,t') e^{-(\gamma_1 + \gamma_2 + \gamma_a)(t-t')/2} + \mu_{43}^2 \int_0^t dt' (\rho_{44} - \rho_{33}) \mathcal{E}(z,t') e^{-\gamma_{34}(t-t')} \right]. \quad (\text{B.11})$$

The first term in the brackets on the right hand side of Eq. (B.11) has already been considered in Appendix A so that it is only necessary to consider the changes induced by the addition of a system with states 4 and 3.

For small \mathcal{E} (linear gain region) ρ_{44} and ρ_{33} are determined by the equations

$$\rho_{44} \approx \Lambda(z) \delta\left(t - \frac{z}{c}\right) - \gamma_4 \rho_{44},$$

$$\rho_{33} \approx \gamma_3 \rho_{33},$$

so that

$$\rho_{44} \approx \Lambda(z) e^{-\gamma_4 \xi} \theta(\xi), \quad (\text{B.12})$$

$$\rho_{33} = \rho_{33}(0) e^{-\gamma_3 t}, \quad (\text{B.13})$$

where $\xi = t - \frac{z}{c}$ and $\theta(\xi)$ is the unit step function. At time $t = 0$ there is no population in the state 3 so $\rho_{33}(0) = 0$ and $\rho_{33}(t) = 0$. The second term in the brackets in equation (B.11) therefore becomes

$$\begin{aligned} & \mu_{43}^2 \int_0^t d\tau' e^{-\gamma_{34}(t-t')} \mathcal{E}(z, t') \theta(\xi) \\ &= \mu_{43}^2 e^{-\gamma_4 \xi} \theta(\xi) \int_0^\xi d\tau e^{\frac{\gamma_4 - \gamma_a}{2} \tau} \mathcal{E}(z, t-\tau) \\ &= \mu_{43}^2 e^{-\gamma_4 \xi} \mathcal{E}(z, t) \theta(\xi) \int_0^\xi d\tau e^{\frac{\gamma_4 - \gamma_3}{2} \tau}. \end{aligned}$$

The remaining steps are identical with those of Appendix A so they will not be repeated. The final solution is

$$\mathcal{E}^2(z', \xi) = \mathcal{E}^2(0, \xi) \exp \left\{ \int_0^{z'} r(x) dx F(\xi) + \int_0^{z'} r_c(x) dx F_c(\xi) \right\}, \quad (\text{B.14})$$

where $r(x)$ and $F(\xi)$ are defined after Eq. (A.13) and by Eq. (A.17) respectively and

$$r_c(x) = \frac{\Lambda(x) \mu_{43}^2 N v}{c \epsilon_0 h}, \quad (\text{B.15})$$

$$F_c(\xi) = e^{-\gamma_4 \xi} \theta(\xi) \int_0^\xi d\tau e^{\frac{\gamma_4 - \gamma_3}{2} \tau}, \quad (\text{B.16})$$

The only difference between Eq. (B.14) and (A.17) (pumping with a 16.997 KeV instead of a 8.95 KeV beam) is the gain coefficient which is increased by an amount

$$\frac{N\mu_{43}^2 v}{c \epsilon_0 h} F_c(\xi) \int_0^{z'} f(x) dx$$

where $\Lambda(z) = \Lambda f(z)$. The k_{α_1} radiation still increases exponentially as the pumping rate Λ is increased.

This model suffers from the same problems that were listed in Appendix A. It suffers from the further problem that states 4 and 3 should be replaced by a distribution of states.

Comparison of the predictions of this model and the experimental results awaits completion of the experiment.

APPENDIX C

In order to comprehend the results of the Das Gupta experiments,¹ it is necessary to fully understand how a crystalline lattice affects the processes that occur within a single atom in the crystal. For example, an electromagnetic wave in a crystal is more likely to propagate in certain directions than in others. This means that any process where the atom emits radiation will be affected by the crystal. Of particular importance for the Das Gupta experiment¹ is the effect of the crystal on the spontaneous and stimulated emission of an atom in that crystal.

Lee and Lee³ have investigated the spontaneous decay rate of an excited atom in a crystal made of identical atoms, resonant with the excited atom. This is not the case that occurs when the excited atom is created by the removal of one of the inner shell electrons. In order to treat this particular case, we have considered the spontaneous emission of an initially excited two-level atom in a crystalline array of two-level atoms. The initially unexcited atoms are identical with each other but not resonant with the excited atom. The excited atom is assumed to have a Bohr frequency ω_0 and to be located at \vec{x} . The remaining $N-1$ atoms have a Bohr frequency ω_j and are located at positions \vec{x}_j , $j = 1, 2, \dots, N-1$.

The equation of motion of the probability amplitudes for the various states of the system are given by

$$(i \frac{\partial}{\partial t} - \omega_0) a(t) = \epsilon \sum_{\vec{k}} A_{\vec{k}}^* e^{i\vec{k} \cdot \vec{x}} b_{\vec{k}}(t), \quad (C.1)$$

$$(i \frac{\partial}{\partial t} - \omega_j) a_j(t) = \epsilon \sum_{\vec{k}} B_{\vec{k}} e^{i\vec{k} \cdot \vec{x}_j} b_{\vec{k}}(t), \quad (C.2)$$

$$(i \frac{\partial}{\partial t} - \omega_{\vec{k}}) b_{\vec{k}}(t) = \epsilon \left[\begin{aligned} & \epsilon A_{\vec{k}} e^{-i\vec{k} \cdot \vec{x}} a(t) \\ & + \sum_{j=1}^{N-1} \epsilon B_{\vec{k}} e^{-i\vec{k} \cdot \vec{x}_j} a_j(t) \end{aligned} \right], \quad (C.3)$$

with the initial condition $a(0) = 1$, $a_j(0) = b_{\vec{k}}(0) = 0$ where $\epsilon A_{\vec{k}}$ and $\epsilon B_{\vec{k}}$ are coupling constants proportional to dipole matrix elements for the initially excited atom and unexcited atoms respectively, $a(t)$ is the probability amplitude that the initially excited atom is excited at time t , all other atoms are in their ground state and there are no photons in the radiation field; $b_{\vec{k}}$ is the probability amplitude that there is one photon with wave vector \vec{k} in the radiation field and all atoms are in their ground state, and a_i is the amplitude for the i th atom to be excited, no photons and all other atoms in their ground state. The quantity $\epsilon A_{\vec{k}}$ is given by

$$\epsilon A_{\vec{k}} = - \left(\frac{2\pi\hbar\omega_{\vec{k}}}{V} \right)^{\frac{1}{2}} \frac{\langle + | \vec{p}/m | \uparrow \rangle}{\hbar\omega_{\vec{k}}}, \quad (C.4)$$

where V is the volume, of the crystal, $| \uparrow \rangle$ is the ground state of the initially excited atom and \vec{p} is the momentum of the electron around the atoms. A similar expression can be written for $\epsilon B_{\vec{k}}$.

Approximate solutions for Eqs. (C.1)-(C.3) can be determined with the aid of the perturbation theory of Lindstedt and Poincaré.⁵ In this method the terms secular in t that arise in conventional perturbation theory are used to renormalize the frequencies ω_0 , ω_s , and ω_k . If the renormalized frequencies corresponding to ω_0 , ω_s , and ω_k are denoted as Ω , Ω_j , Ω_k respectively then new time scales can be defined

$$\tau = \Omega t, \quad (C.5)$$

$$\tau_j = \Omega_j t, \quad (C.6)$$

$$\tau_k = \Omega_k t, \quad (C.7)$$

so that Eqs. (C.1)-(C.3) become

$$(i\Omega \frac{d}{d\tau} - \omega_0) a(\tau) = \epsilon \sum_{\vec{k}} A_{\vec{k}}^* e^{i\vec{k} \cdot \vec{x}} b_{\vec{k}}(\tau_k), \quad (C.8)$$

$$(i\Omega_j \frac{d}{d\tau} - \omega_s) a_j(\tau_j) = \epsilon \sum_{\vec{k}} B_{\vec{k}}^* e^{i\vec{k} \cdot \vec{x}_j} b_{\vec{k}}(\tau_k), \quad (C.9)$$

$$(i\Omega_k \frac{d}{d\tau_k} - \omega_k) b_{\vec{k}}(\tau_k) = \epsilon \left[A_{\vec{k}} e^{-i\vec{k} \cdot \vec{x}} a(\tau) + \sum_{j=1}^{N-1} B_{\vec{k}} e^{-i\vec{k} \cdot \vec{x}_j} a_j(\tau_j) \right]. \quad (C.10)$$

The renormalized frequencies along with the amplitudes are then expanded in powers of ϵ as

$$\Omega = \omega_0 + \epsilon \Omega^{(1)} + \epsilon^2 \Omega^{(2)} + \dots, \quad (C.11)$$

$$\Omega_j = \omega_s + \epsilon \Omega_j^{(1)} + \epsilon^2 \Omega_j^{(2)} + \dots, \quad (C.12)$$

$$\Omega_k = \omega_k + \epsilon \Omega_k^{(1)} + \epsilon^2 \Omega_k^{(2)} + \dots, \quad (C.13)$$

$$a = a^{(0)} + \epsilon a^{(1)} + \epsilon^2 a^{(2)} + \dots, \quad (C.14)$$

$$a_j = a_j^{(0)} + \epsilon a_j^{(1)} + \epsilon^2 a_j^{(2)} + \dots, \quad (C.15)$$

$$b_{\vec{k}} = b_{\vec{k}}^{(0)} + \epsilon b_{\vec{k}}^{(1)} + \epsilon^2 b_{\vec{k}}^{(2)} + \dots \quad (C.16)$$

When Eqs. (C.11)-(C.16) are substituted into Eqs. (C.8)-(C.10) and the coefficients of like powers of ϵ are equated on the following infinite set of first order differential equations arises:

$$\epsilon^0 : \quad (i \frac{d}{d\tau} - 1) a^{(0)} = 0 , \quad (C.17)$$

$$(i \frac{d}{d\tau_j} - 1) a_j^{(0)} = 0 , \quad (C.18)$$

$$(i \frac{d}{d\tau_k} - 1) a_k^{(0)} = 0 , \quad (C.19)$$

$$\begin{aligned} \epsilon^n : \quad \omega_0 (i \frac{d}{d\tau} - 1) a^{(n)}(\tau) &= \sum_{\vec{k}} A_{\vec{k}}^* b_{\vec{k}}^{(n-1)}(\tau_k) e^{i\vec{k} \cdot \vec{x}} \\ &- i \sum_{m=0}^{n-1} \Omega^{(n-m)} \frac{d}{d\tau} a^{(m)}(\tau) , \end{aligned} \quad (C.20)$$

$$\begin{aligned} \omega_j (i \frac{d}{d\tau_j} - 1) a_j^{(n)}(\tau_j) &= \sum_{\vec{k}} B_{\vec{k}}^* b_{\vec{k}}^{(n-1)}(\tau_k) e^{i\vec{k} \cdot \vec{x}_j} \\ &- i \sum_{m=0}^{n-1} \Omega_j^{(n-m)} \frac{d}{d\tau_j} a_j^{(m)}(\tau_j) , \end{aligned} \quad (C.21)$$

$$\begin{aligned} \omega_k (i \frac{d}{d\tau_k} - 1) b_k^{(n)}(\tau_k) &= A_{\vec{k}} e^{-i\vec{k} \cdot \vec{x}_a} a^{(n-1)}(\tau) \\ &+ \sum_{j=1}^{N-1} B_{\vec{k}} e^{-i\vec{k} \cdot \vec{x}_j} a_j^{(n-1)}(\tau_j) \\ &- i \sum_{m=0}^{n-1} \Omega_k^{(n-m)} \frac{d}{d\tau_k} b_k^{(m)}(\tau_k) , \end{aligned} \quad (C.22)$$

where $n = 1, 2, 3, \dots$

The solutions of Eqs. (C.17)-(C.18) are then

$$a^{(0)} = C e^{-i\tau} \quad (C.23)$$

$$a_j^{(0)} = C_j e^{-i\tau_j} \quad (C.24)$$

$$b_{\vec{k}}^{(0)} = D_{\vec{k}} e^{-i\tau_k} \quad (C.25)$$

where C , C_j , $D_{\vec{k}}$ are constants to be determined from the boundary conditions. Substitution of Eqs. (C.23) and (C.24) into Eq. (C.20) for the case when $n = 1$ gives:

$$\omega_0 i \frac{d}{d\tau} (a^{(1)}(\tau) e^{i\tau}) = \sum_{\vec{k}} A_{\vec{k}} D_{\vec{k}} e^{i(\tau-\tau_k)} e^{i\vec{k}\cdot\vec{x}} - C\Omega^{(1)}. \quad (C.26)$$

When this equation is integrated the second term on the right hand gives rise to secular divergent behaviour in τ . In order to remove this problem the solution is renormalized by choosing Ω

$$\Omega^{(1)} = 0.$$

Equation (C.26) can now be integrated to give:

$$a^{(1)}(\tau) = \frac{\Omega}{\omega_0} \sum_{\vec{k}} \frac{A_{\vec{k}}^* D_{\vec{k}}}{\Omega_{\vec{k}} - \Omega} e^{-i\tau_k} e^{i\vec{k}\cdot\vec{x}}.$$

Likewise Eqs. (C.21) and (C.22) for the case when $n = 1$ can be integrated to give

$$\Omega_{\vec{k}}^{(1)} = \Omega_j^{(1)} = 0 ,$$

$$a_j^{(i)}(\tau_j) = \frac{\Omega_j}{\omega_0} \sum_{\vec{k}} \frac{B_{\vec{k}}^* D_{\vec{k}}}{\Omega_{\vec{k}} - \Omega_j} e^{i\vec{k} \cdot \vec{x}_j} e^{-i\tau_{\vec{k}}} ,$$

$$b_{\vec{k}}^{(1)}(\tau_{\vec{k}}) = \frac{\Omega_{\vec{k}}}{\omega_{\vec{k}}} \left[\frac{A_{\vec{k}} C_{\vec{k}}}{\Omega - \Omega_{\vec{k}}} e^{-i\vec{k} \cdot \vec{x}} e^{-i\tau} \right. \\ \left. + B_{\vec{k}} \sum_{j=1}^{N-1} \frac{C_j}{\Omega_j - \Omega_{\vec{k}}} e^{-i\vec{k} \cdot \vec{x}_j} e^{-i\tau_j} \right] .$$

In this manner, Eqs. (C.20)-(C.22) can be integrated for $n \geq 2$. For each value of n the known quantities are substituted into the equations which are then integrated. Those terms that are secular are eliminated from the equation by setting their sum equal to zero. This process renormalizes the frequency.

Rather than write down all the intervening steps, it is appropriate to skip to the final result because the intermediate steps are straight forward but tedious. Because the object of this exercise is to determine the lifetime of an excited atom in a nonresonant crystal, only the frequency of the solution of $a(\tau)$ is important. This frequency to fourth order in ϵ is

$$\Omega - \Omega_s = - \frac{\Omega}{\omega_s} \sum_{\vec{k}} \sum_{j=1}^{N-1} \frac{\Omega_{\vec{k}} \Omega_j}{\omega_{\vec{k}}^2} \frac{\epsilon^4 |A_{\vec{k}}|^2 |B_{\vec{k}}|^2}{(\Omega_j - \Omega_{\vec{k}})(\Omega - \Omega_{\vec{k}})^2} \\ + \epsilon^4 \sum_{\vec{k}} \sum_{\vec{k}'} \sum_{j=1}^{N-1} \frac{\Omega_j}{\omega_s} \frac{\Omega_{\vec{k}}}{\omega_{\vec{k}}} \frac{\Omega_{\vec{k}'}}{\omega_{\vec{k}'}} \frac{A_{\vec{k}}^* A_{\vec{k}'}, B_{\vec{k}}^* B_{\vec{k}'}}{(\Omega_{\vec{k}} - \Omega)(\Omega_{\vec{k}'} - \Omega)(\Omega_j - \Omega)} e^{i(\vec{k} - \vec{k}') \cdot (\vec{x} - \vec{x}_j)} , \quad (C.27)$$

where Ω_s is the renormalized frequency that would occur if the atom were in free space. In order to simplify Eq. (C.27) the renormalized frequencies on the right hand side can be set equal to their values when the coupling constants are zero

$$\Omega \approx \omega_0, \quad (C.28)$$

$$\Omega_j \approx \omega_s, \quad (C.29)$$

$$\Omega_k \approx \omega_k.$$

With these approximations Eq. (C.27) becomes

$$\begin{aligned} \Omega - \Omega_s \approx & - (N-1) \sum_{\vec{k}} \frac{\omega_0}{\omega_k} \frac{\epsilon^4 |A_{\vec{k}}|^2 |B_{\vec{k}}|^2}{(\omega_s - \omega_k)(\omega_0 - \omega_k)^2} \\ & + \epsilon^4 \sum_{\vec{k}} \sum_{\vec{k}'} \frac{A_{\vec{k}}^* B_{\vec{k}'} B_{\vec{k}} B_{\vec{k}'}^*}{(\omega_k - \omega_0)(\omega_{k'} - \omega_0)(\omega_s - \omega_0)} \sum_{j=1}^{N-1} e^{i(\vec{k} - \vec{k}') \cdot (\vec{x} - \vec{x}_j)}. \end{aligned} \quad (C.31)$$

The summation over j that occurs in the last term of Eq. (C.31) can now be performed

$$\sum_{j=1}^{N-1} e^{i(\vec{k} - \vec{k}') \cdot (\vec{x} - \vec{x}_j)} \approx (N-1) \sum_{\vec{q}} \delta_{\vec{k} - \vec{k}', \vec{q}}, \quad (C.32)$$

where \vec{q} is a reciprocal lattice vector. The argument of the delta function in Eq. (C.32) simply states the conditions for Bragg scattering of the radiation in a crystal.

Equations (C.31) and (C.32) represent the current state of these calculations. Future work will include division of Eq. (C.31) into its real and imaginary parts (separate the level shift from the linewidth) and approximate evaluation of the sums.

The work presented in this report was performed by M. O. Scully, Professor of Physics and Optical Sciences, University of Arizona; G. T. Moore and J. K. McIver, Research Physicists, University of Arizona.

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