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Theory of opacity from two-photon absorption processes

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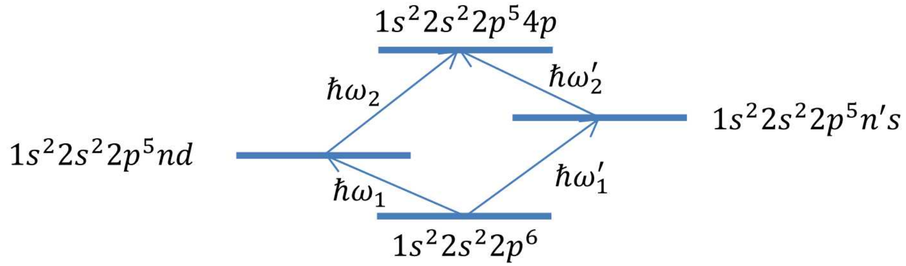
Introduction

We describe recent research on plasma opacity produced by two-photon absorption.

$$\sum_{n,n'}$$



- **Principal motivation:** series of foil transmission experiments performed by J. Bailey et al. on the Z machine in the Sandia Laboratory: higher measured opacity for hot Fe foil samples than predicted by several well-known opacity theory codes.

The mystery arises in a context of uncertainty in modeling the Solar interior.

- Quantum theory of two-photon emission/absorption published by Goeppert-Mayer in 1931 and applied to emission from metastable hydrogen in interstellar space by Breit and Teller.

Two-photon cross-sections are obtained using Fermi's "Golden Rules" for quantum perturbation theory.

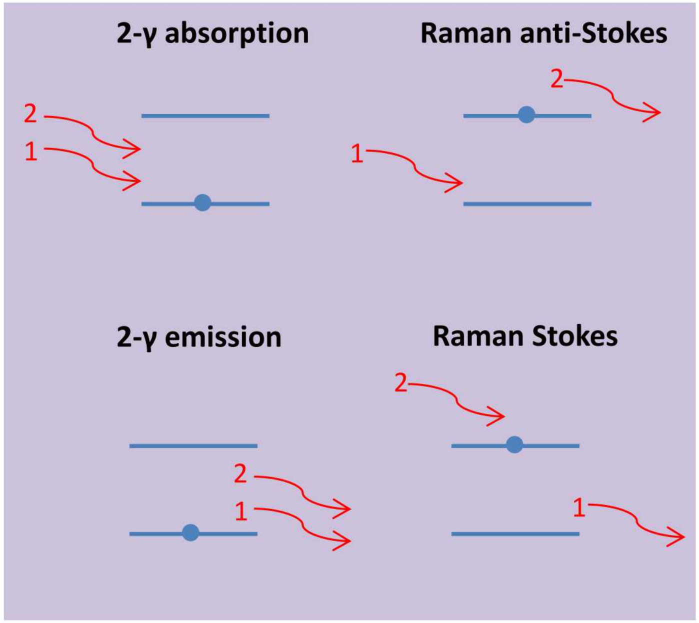
- We investigate the two-photon process in which **one photon comes from a backlighter radiation source** and **the other photon comes from the ambient plasma**.
- The fact that the two photons are not identical greatly increases this process rate.

M. Goeppert-Mayer, Ann Phys 9, 273 (1931).
G. Breit and E. Teller, ApJ **91**, 215 (1940).

Introduction

■ Calculation of two-photon opacity is challenging because four radiative processes - **absorption**, **emission** and **two Raman effects (Stokes and anti-Stokes)** - occur at each photon energy.

- Necessary to sum over various classes of intermediate states, including different orders of photo-absorption and electron excitation;
- Necessary to evaluate angular averages over **radiation field**;
- Integrals over the continuum states are **singular integrals**.



■ Atomic data for the calculations is generally available, although:

- **free-free dipole matrix elements** present special difficulties,
- high density of the experiments raises an issue of plasma effects on the matrix elements.

We describe our own evaluation of these matrix-elements.

A new sum-rule is directly aimed at the two-photon opacity.

Our calculations (based on second-order perturbation theory) are tested by a second computational approach, the numerical solution of the time-dependent Schrödinger equation for an ion interacting with high-frequency electromagnetic fields ("AC Stark Effect" model). The latter includes Raman processes and provides an independent approximate calculation of one-, two- and even three-photon cross-sections.

The Z iron experiment

- Dipole matrix element: $-eFD_{n\ell}^{n'\ell'}$ where $D_{n\ell}^{n'\ell'}=\int_0^\infty R_{n\ell}(r)\,R_{n'\ell'}(r)r^3dr.$

- (Very) simplified two-photon matrix element: $-e^2F^2\sum_{n'',\ell''}D_{n\ell}^{n''\ell''}\frac{1}{E_{n\ell}-E_{n''\ell''}-\hbar\omega}D_{n''\ell''}^{n'\ell'}.$

- Two-photon effect becomes of the same order of one-photon contribution if $eFD\approx e^2F^2\frac{R\times R}{\Delta E}\times(2\pi\alpha)$ (R is a general notation for $D_{n\ell}^{n'\ell'}$).

- Since $\Delta E\approx 2Ryd\frac{Z_{\mathrm{eff}}^2}{n^3}$ and (If n and n' large but close to each other) and D$\approx\frac{3}{2}n^2\frac{a_0}{Z_{\mathrm{eff}}}$, we finally get

$$F\approx\frac{Z_{\mathrm{eff}}^3e}{12\pi^2\alpha\epsilon_0a_0^2n^5}\approx Z_{\mathrm{eff}}^3\frac{7.5\times10^{12}}{n^5}\approx\frac{2.5\times10^{16}}{n^5}\ \mathrm{V/m}$$

and the corresponding flux is $\Phi_{\mathrm{min}}=c\epsilon_0\frac{F^2}{2}\approx\frac{8.5\times10^{25}}{n^{10}}\mathrm{W/cm^2}$.

- For the SNL experiment, backlighter temperature is $T_{\mathrm{BL}}\approx 350\ eV$ and the dilution factor $f_d\approx 0.13$.

The flux on the sample is $\sigma T_{\mathrm{eff}}^4\approx 0.13\times\sigma T_{\mathrm{BL}}^4\approx 2\times10^{14}\ \mathrm{W/cm^2}$ and $\Phi_{\mathrm{min}}=\sigma T_{\mathrm{eff}}^4$ implies $\boldsymbol{n\geq 15}$ …

But, in the conditions of the experiment: $n_e\approx 3.1\ \ 10^{22}\ \mathrm{cm^{-3}}$, due to density effects, the last populated subshell corresponds to $n=8$ …

J.-C. Pain, High Energy Density Phys. **26**, 23 (2018).

R. M. More's relevant criticism

- One should consider **two photons of different energies** $\hbar\omega_1, \hbar\omega_2 \rightarrow \sigma(\omega_1, \omega_2)$

$\omega_1 = \omega_2$ occurs only by accident and makes a tiny contribution.

- M. Kruse made a 1-color calculation (RPHDM 2016) and found $\sigma(\omega_1, \omega_2)$ was too small.

He calculated $\sigma(\omega_1, \omega_1)$ but agreed he should find a way to do the two-color cross-sections.

- With the AC code, R M More calculated $\sigma(\omega_1, \omega_1)$ for $2s \rightarrow 4d$ with $\hbar\omega_1$=586.14 eV and found 5.62 10^{-54} cm^4 s eV, which is also too small.

Everybody agree it's too small, but it's not the right process!

- Photon ω_1 is from the backlighter, the other photon is from the plasma or from the backlighter.

Total photon energy is constrained : $\hbar\omega_1 + \hbar\omega_2 = \Delta E = E_{\text{final}} - E_{\text{initial}}$.

- For any $\hbar\omega_1$ ($< \Delta E$), there can be a second photon that has the right energy $\hbar\omega_2 = \Delta E - \hbar\omega_1$.
- It is a continuous absorption, even for bound-bound transitions.
- It should be compared to the low-opacity gaps between one-photon lines.

- The opacity of attenuation of photon 1 is proportional to integral $\int \sigma(\omega_1, \omega_2) c n(\omega_2) g(\omega_2) d\omega_2$

The integral is much larger than the cross-section for two identical photons.

- Cross-section per electron and per process: $\sigma(\omega_1, \omega_2) \approx 10^{-54}$ cm^{4}s eV (from R. M. More's AC Stark code for Fe^{16+}). M. S. Pindzola and J. Colgan got the same order of magnitude for $h\omega_1 = h\omega_2$.

- Photon energy $\hbar\omega_{\text{max}} \sim 5k_B T$, dilution factor $f_d \sim 0{,}1$, $k_B T = 200$ eV $\rightarrow$ $\sigma T^4 \sim 1.6\ 10^{14}$ W/cm^2.

- Photon flux ($h\nu_2$): $\Phi_2 = f_d \frac{\sigma T^4}{\hbar\omega_{\text{max}}} \approx 10^{29}$ cm^{-2}s^{-1}eV^{-1} for $T_R \approx 200$ eV.

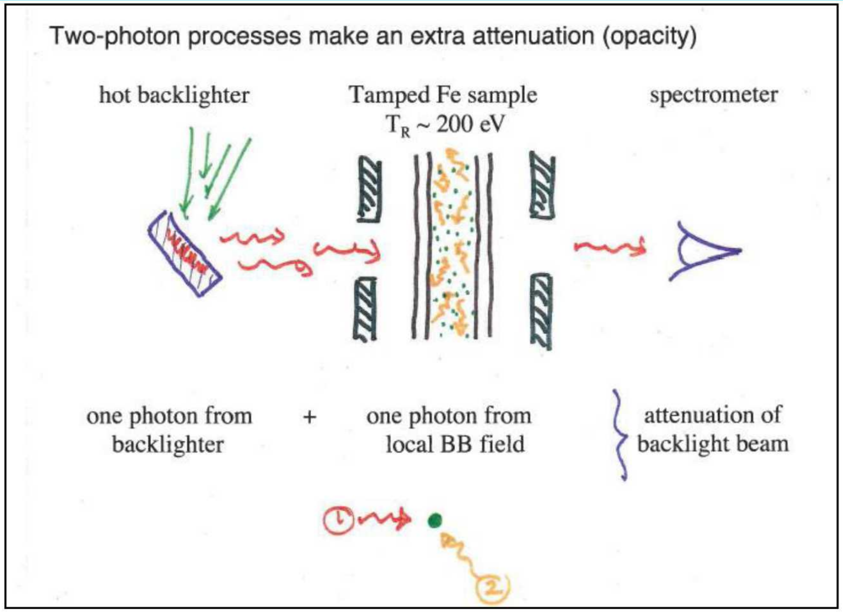
N_ν: Number of eV-size photon energy groups in $[0, 5T_R] \approx 10^3$.
N_e: Number of active electrons $(2s, 2p)$ in Ne-like Fe: ≈ 8.
N_{proc}: Number of processes (i.e., final states): $\approx 10^2$.

Examples: $2s \rightarrow 3d, 2s \rightarrow 4d, 2s \rightarrow \epsilon d$ (continuum final states), $2\text{p} \rightarrow 4f, etc.$

Estimate: $\sigma \Phi_2 N_\nu N_e N_{proc} \approx 10^{-54} \times 10^{29} \times 10^3 \times 8 \times 10^2 \approx 8\ 10^{-20}$ cm^2 **which is enough extra opacity to understand the Sandia Fe experiment!**

In addition, as will be shown in figure 1, the cross-section is sometimes much larger (even 6 orders of Magnitude larger, see Fig. 1)!

We all made simplifying assumptions…



R. M. More, S. B. Hansen and T. Nagayama, High Energy Density Phys. **24**, 44 (2017):
- Calculations shown in Santa Barbara in November 2016 gave an opacity change that was too large and had odd oscillations accross the range of the measured spectrum.
- left out the bound-free and free-free contributions because the free-free matrix elements were unreliable. Density effects were not included.

J.-C. Pain, High Energy Density Phys. **26**, 23 (2018):
- omitted two-color two-photon absorption.
- omitted intermediate states in the continuum.
- did not consider two-photon photo-ionization. They involve final states in the continuum and are even more important when there is continuum lowering.
- did not consider Raman (Stokes and Anti-Stokes) processes. They contribute to opacity too.

Free-free matrix elements for two-photon opacity

Five ways to compute the free-free matrix elements for two-photon opacity $R_{E,\ell}^{E',\ell'}$:

1. B. Gao and A. Starace[1] published 8 calculations using an original method of « complex contour rotation » limited to pure Coulomb potential (H-like ions), but that can be scaled using effective charges. No density effets included. No information about behavior at $E = E'$ (studied by V. Veniard and B. Piraux[2]).

2. Analytic continuation of the Gordon formula for bound-bound matrix elements. Limited to H-like ions. No idea how to include density effects. Results are good near the $E = E'$ singularity but $R_{E,\ell}^{E,\ell'}$ diverges.

3. Saddle-point method and WKB wavefunctions. Described by More and Warren[3] for bound-bound and bound-free transitions, but difficult to extend to free-free transitions.

4. Numerical integration. Calculations started using confluent hypergeometric function at small radius. Enhanced Simpson rule is used to form the continuum wavefunctions. Agrees with methods 1 and 2 to a fraction of %. Can be extended to include density effects.

5. Acceleration formula with numerical WKB wavefunctions. Differences with method 4 around the inner turning point.

[1]B. Gao and A. Starace, Numerical Methods for free-free radiative transition matrix elements, University of Nebraska Digital Commons (1987).
[2]V. Veniard and B. Piraux, Phys. Rev. A **41**, 4019 (1990).
[3]R. More and K. H. Warren, Ann. of Phys. **207**, 282 (1991).

Free-free matrix elements for two-photon opacity

Compare Gao-Starace, Analytic continuation and numerical wave-functions

				Gao-Starace	Analytic continuation	Numerical integral
E = .016	$l = 0$	E' = .059	$l' = 1$	$R_{GS} = 111.060$	$R_{AC} = 111.064$	$R_{NI} = 111.0906$
E = .016	$l = 1$	E' = .059	$l' = 0$	$R_{GS} = 57.943$	$R_{AC} = 57.9434$	$R_{NI} = 58.0109$
E = .016	$l = 1$	E' = .059	$l' = 2$	$R_{GS} = 114.240$	$R_{AC} = 114.243$	$R_{NI} = 114.229012$
E = .016	$l = 2$	E' = .059	$l' = 1$	$R_{GS} = 33.414$	$R_{AC} = 33.4126$	$R_{NI} = 33.479428$
E = .100	$l = 0$	E' = .700	$l' = 1$	$R_{GS} = 1.29930$	$R_{AC} = 1.29927$	$R_{NI} = 1.299249$
E = .100	$l = 1$	E' = .700	$l' = 0$	$R_{GS} = .35695$	$R_{AC} = .356944$	$R_{NI} = .356911$
E = .100	$l = 1$	E' = .700	$l' = 2$	$R_{GS} = .72339$	$R_{AC} = .723395$	$R_{NI} = .723416$
E = .100	$l = 2$	E' = .700	$l' = 1$	$R_{GS} = .08783$	$R_{AC} = .0878189$	$R_{NI} = .087823$

Method 2	E = 0.5	$A(E,s \to E\text{-}\delta E,p) =$ .22071	extrap to $A(E,s \to E,p) =$	.2234755
Method 4		.221002	evaluation at E = E"	.226056
Method 2	E = 0.6	$A(E,s \to E\text{-}\delta E,p) =$ .23097	extrap to $A(E,s \to E,p) =$	.233652
Method 4		.231341	evaluation at E = E"	.235935
Method 2	E = 0.7	$A(E,s \to E\text{-}\delta E,p) =$ .23924	extrap to $A(E,s \to E,p) =$	.241816
Method 4		.239659	evaluation at E = E"	.243863
Method 2	E = 0.8	$A(E,s \to E\text{-}\delta E,p) =$ .24605	extrap to $A(E,s \to E,p) =$	.248521
Method 4		.246496	evaluation at E = E"	.250375
Method 2	E = 0.9	$A(E,s \to E\text{-}\delta E,p) =$ .251763	extrap to $A(E,s \to E,p) =$	.254131
Method 4		.252225	evaluation at E = E"	.255822

Two-photon perturbation theory vs. AC Stark code

- **Bridge code**: second-order perturbation theory.
- **AC Stark code**: solves time-dependent Schrödinger equation for Fe^{16+} ion subject to two overlapping pulses of X rays.

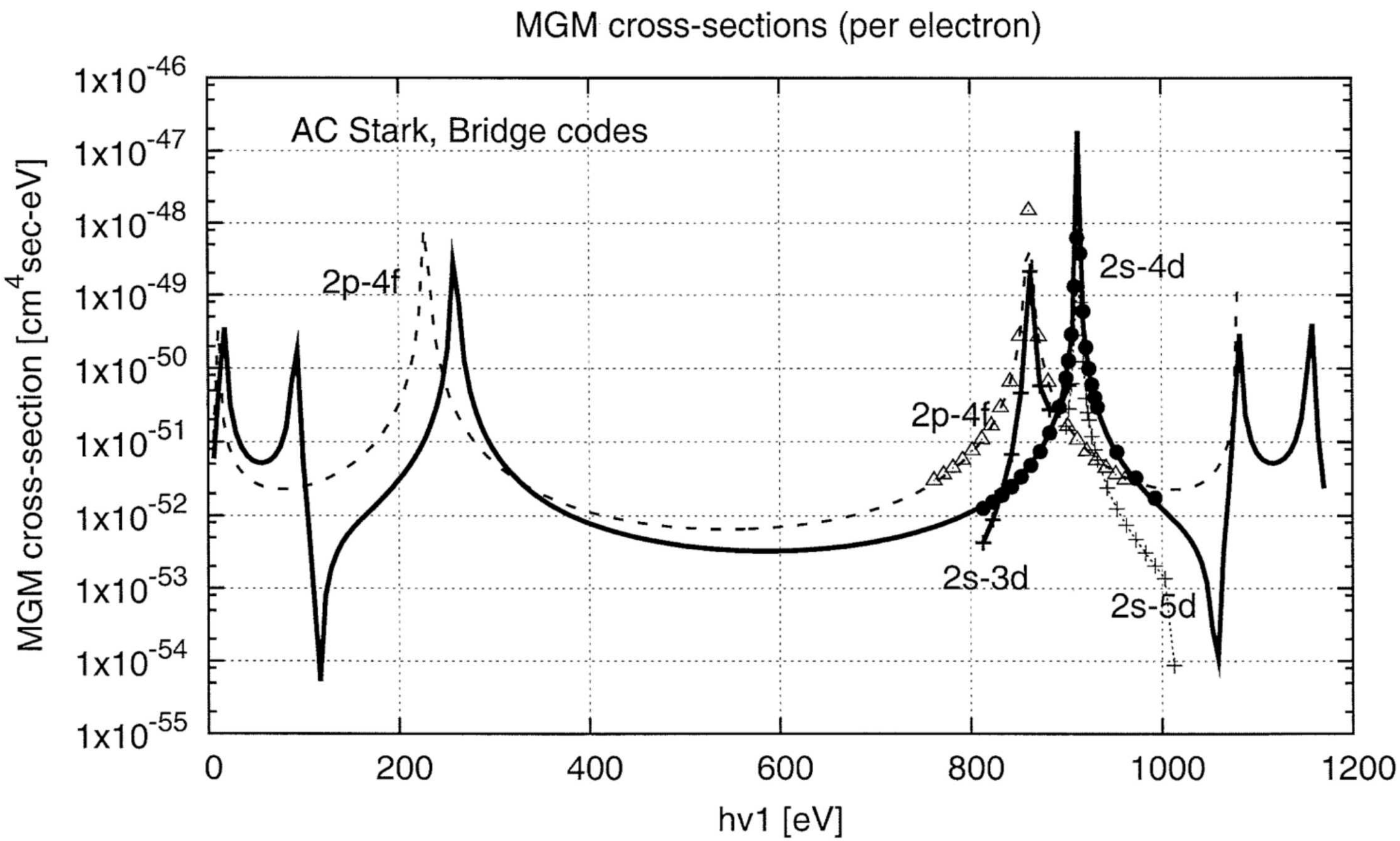


Figure 1: solid and dashed lines: Bridge code, points: AC Stark code. The same list of states, energies and matrix elements were used for the two codes.

MGM cross-section per electron ($2s, 2p \rightarrow n = 3$ to 7)

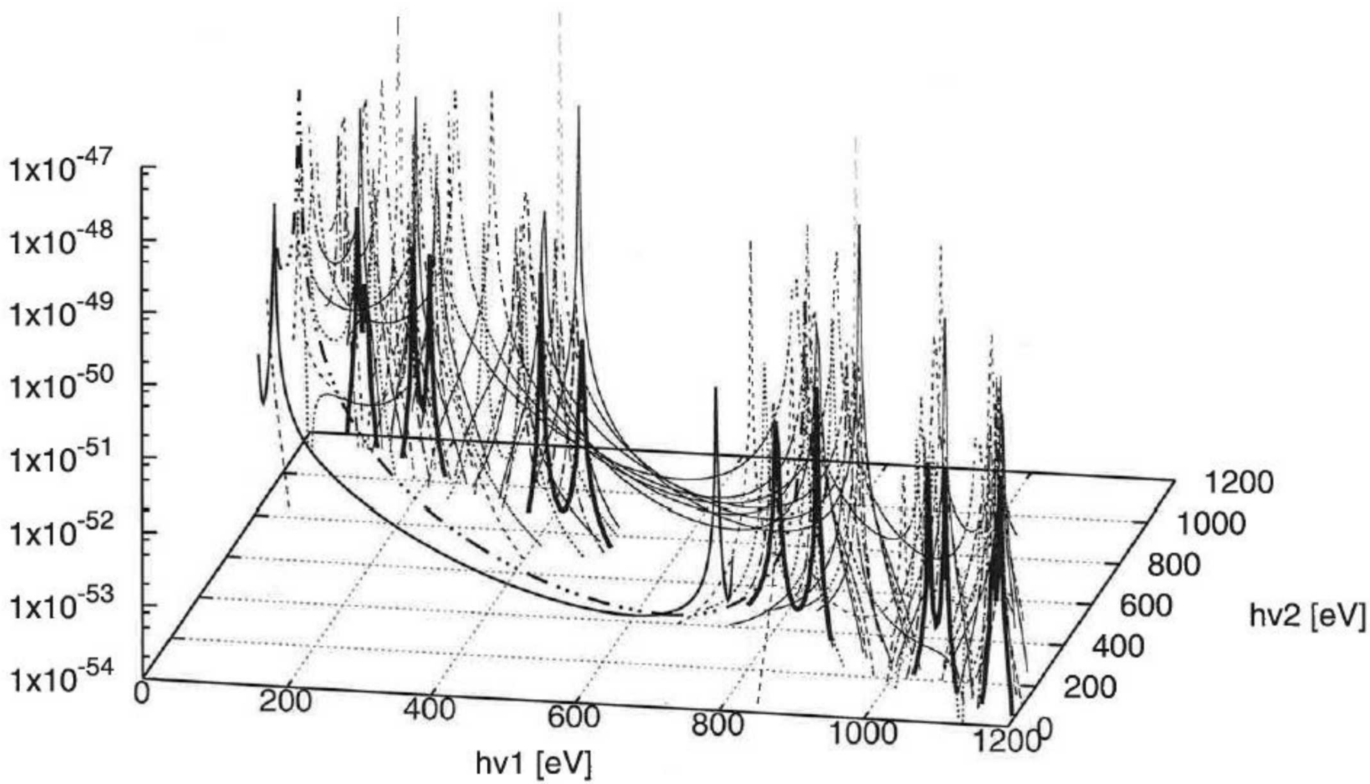
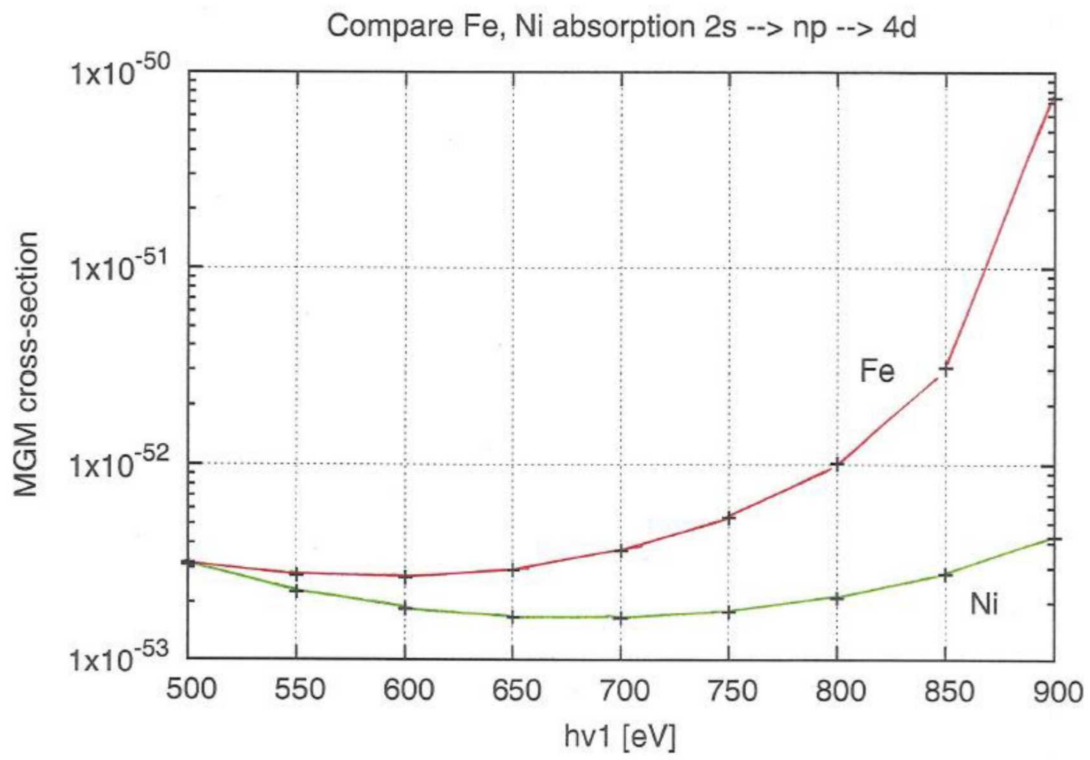


Figure 2: Bridge code for 19 transitions from 2s, 2p states of Ne-like Fe.

Why is there an anomaly for Fe but not for Ni?

■ Since the energies are ~200 eV larger for Ni than for Fe, the cross-section and the population from the Bose-Einstein factor are smaller. **The 2-photon process in Ni is 10 times smaller than for Fe**. That helps to understand why it is not visible in the experiments that have been done. At a higher temperature, it might be visible.



$$\sigma^{MGM} = 8\pi^3\left(\frac{e^2}{\hbar c}\right)^2 \frac{\hbar}{(\hbar\omega_1)(\hbar\omega_2)} \left| \sum_n \frac{\delta E_{in} R_{in}\ \delta E_{nf} R_{nf}}{E_i - E_n} \right|^2 \Gamma_{ang}\ I(\hbar\omega_1 + \hbar\omega_2)$$

A two-photon sum rule to check the calculations

- Let us consider the Hamiltonian

$$H=\frac{p^2}{2m}+V(r)$$

and define the suite of successive commutations of r with H :

$$C^{(0)}=r \quad ; \quad C^{(1)}=[H,r]$$
$$C^{(2)}=[H,[H,r]]=[H,C^{(1)}]$$
$$C^{(3)}=[H,[H,[H,r]]]=[H,C^{(2)}]$$

i.e., in the general case

$$C^{(k)}=[H,C^{(k-1)}]$$

It is easy to see that

$$\langle m|C^{(k)}|t\rangle=(E_m-E_t)^k\langle m|r|t\rangle$$

where $|m\rangle$ is an eigenstate (for instance $|n\ell\rangle$) so that $H|m\rangle=E_n|m\rangle$. We have

$$\langle m|[r,C^{(k)}]|q\rangle=\langle m|rC^{(k)}|q\rangle-\langle m|C^{(k)}r|q\rangle=\sum_t\langle m|r|t\rangle\langle t|C^{(k)}|q\rangle-\sum_t\langle m|C^{(k)}|t\rangle\langle t|r|q\rangle$$
$$=\sum_t\langle m|r|t\rangle\left(E_t-E_q\right)^k\langle t|r|q\rangle-\sum_t(E_m-E_t)^k\langle m|r|t\rangle\langle t|r|q\rangle$$

A two-photon sum rule to check the calculations

- In order to obtain a sum rule, we have to evaluate $\langle m|[r, C^{(k)}]|q\rangle$. For k=1 :

$$C^{(1)} = -\frac{1}{2m}[r, p^2] = -i\hbar\frac{p}{m}$$

$$\langle m|[r, C^{(1)}]|q\rangle = \langle m|[r, [H, r]]|q\rangle = \left\langle m\left|\left[r, \frac{i\hbar}{m}p\right]\right|q\right\rangle = -\frac{(i\hbar)^2}{m}\langle m|q\rangle = \frac{\hbar^2}{m}\delta_{m,q}$$

which yields

$$\sum_t \langle m|r|t\rangle(E_t - E_q)\langle t|r|q\rangle - \sum_t (E_m - E_t)\langle m|r|t\rangle\langle t|r|q\rangle = \frac{\hbar^2}{m}\delta_{m,q}$$

Or

$$2\sum_t \left[E_t - \frac{1}{2}(E_q + E_m)\right]\langle m|r|t\rangle\langle t|r|q\rangle = \frac{\hbar^2}{m}\delta_{m,q}$$

Changing from vector r ro radial dipole R giving a factor 1/3 for s – p transitions and setting $|m\rangle = |n_1 s\rangle$, $|q\rangle = |n_3 s\rangle$ and $|t\rangle = |n_2 p\rangle$, we obtain

$$2\sum_{n_2} \left[E_{n_2 p} - \frac{1}{2}(E_{n_1 s} + E_{n_3 s})\right]\langle n_1 s|r|n_2 p\rangle\langle n_2 p|r|n_3 s\rangle = \frac{\hbar^2}{m}\delta_{n_1,n_3}$$

■ We were wondering: **does two-photon absorption affect radiative diffusion[2] in stars?**

■ Stellar envelope has normal diffusion $\propto \nabla n_Z$ and radiation-driven diffusion $\propto \kappa_Z \nabla T_R$.

■ To our knowledge, the problem of **acceleration (diffusion) induced by two-photon radiation** was never studied before.

➢ The diffusion current is related to the opacity as an integral over the "out-of-equilibrium" part of the radiation field. For two-photon absorption, there is a similar integral, a little more complicated, which might yield **extra diffusion** due to the two-photon absorption.

■ A phenomenon might be affected by two-photon radiative acceleration: the so-called **"saturation effect[3]".** When matter density increases, the number of ions per volume unit getting higher, the number of available photons likely to yield the acceleration decreases.

➢ For two-color absorption, saturation should be weaker, because of the many possible intermediate states, and the process is less stringent as concerns the photon energy; however, one has to ensure $h\nu_1 + h\nu_2 = h\nu$, which is also a strong constraint, and therefore may increase the saturation…

[1]S. Turcotte, J. Richer, G. Michaud, C. A. Iglesias and F. J. Rogers , ApJ **504**, 539 (1998).
[2]G. Michaud, ApJ **160**, 641 (1970).
[3]G. Alecian and F. LeBlanc, MNRAS **319**, 677 (2000).

Backup

- Unlinked cluster theorem of Many Body Perturbation Theory

- Consider $1s^2 2s^2 2p^6 \rightarrow 1s^2 2s 2p^5 3p 3d$

There are four terms: which electron (2s or 2p) moves first, which photon is absorbed first.

- Energy conservation: $\epsilon_{2s} + \epsilon_{2p} + \hbar\omega_1 + \hbar\omega_2 = \epsilon_{3p} + \epsilon_{3d}$

or $\epsilon_{2p} - \epsilon_{3d} + \hbar\omega_1 = \epsilon_{3p} - \epsilon_{2s} - \hbar\omega_2 = -(\epsilon_{2s} - \epsilon_{3p} + \hbar\omega_2)$

- Channel 1 has 2s going first: $1s^2 2s^2 2p^6 \rightarrow 1s^2 2s 2p^6 3p \rightarrow 1s^2 2s 2p^5 3p 3d$

$$\frac{\langle 3p|\hat{e}_1.\vec{r}|2s\rangle\langle 3d|\hat{e}_2.\vec{r}|2p\rangle}{\epsilon_{2s} - \epsilon_{3p} + \hbar\omega_2} + \frac{\langle 3p|\hat{e}_2.\vec{r}|2s\rangle\langle 3d|\hat{e}_1.\vec{r}|2p\rangle}{\epsilon_{2s} - \epsilon_{3p} + \hbar\omega_1}$$

- Channel 2 has 2p going first: $1s^2 2s^2 2p^6 \rightarrow 1s^2 2s^2 2p^5 3d \rightarrow 1s^2 2s 2p^5 3p 3d$

$$\frac{\langle 3d|\hat{e}_1.\vec{r}|2p\rangle\langle 3p|\hat{e}_2.\vec{r}|2s\rangle}{\epsilon_{2p} - \epsilon_{3d} + \hbar\omega_2} + \frac{\langle 3d|\hat{e}_2.\vec{r}|2p\rangle\langle 3p|\hat{e}_1.\vec{r}|2s\rangle}{\epsilon_{2p} - \epsilon_{3d} + \hbar\omega_1}$$

The four terms cancel!