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Opacity from two-photon processes[☆]

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ABSTRACT

The recent iron opacity measurements performed at Sandia National Laboratory by Bailey and collaborators have raised questions about the completeness of the physical models normally used to understand partially ionized hot dense plasmas. We describe calculations of two-photon absorption, which is a candidate for the observed extra opacity. Our calculations do not yet match the experiments but show that the two-photon absorption process is strong enough to require careful consideration.

1. Introduction

This paper describes the calculation of opacity resulting from two-photon processes in hot dense plasmas. The objective is to understand the Sandia Z-machine experiment published by Bailey et al. in 2015 [1]. The experiments observe an opacity larger than predicted by several well-known opacity theory codes for dense Fe foils at ~ 180 eV temperature. The extra opacity was especially striking in the continuum wavelength range 7–10 Å.

A previous experiment in similar geometry, but at a lower temperature, agreed with theory [1]. The measured opacity is not just larger than predicted by normal opacity theory; it also is larger than the handbook cold opacity in this wavelength range. It has also been suggested that the measured opacity is larger than permitted by the f-sum rule [2] - that is, the sum-rule for dipole-allowed one-photon transitions - but the published measurements do not span a sufficient photon energy-range to evaluate this concern.

The opacity measured in the 180 eV experiments was about 1000 cm^2/gm larger than the opacities predicted by several theoretical models over much of the published frequency range. This difference is roughly equivalent (for Fe) to an extra cross-section of 10^{-19} cm^2 per atom. In the region of line transitions, the difference was not so large and many observed line features agree with the theories.

The calculations in this paper do not agree with the experiment but give an additional two-photon opacity with a similar order-of-magnitude. At this writing, the effects of high density of the target foils are not adequately treated in our calculations, and we conjecture that improved treatment of the density effects on the two-photon process can bring a satisfactory solution to the question. The immediate goal of this paper is to identify the ingredients we consider necessary for a calculation of two-photon opacity.

2. Two-photon emission

Not much has been published about two-photon absorption of X-rays but considerable effort has been invested in understanding the inverse two-photon emission process. Two-photon radiation processes were predicted by Maria Goeppert-Mayer in the 1920's [3]. In the 1930's Breit and Teller calculated two-photon emission from the metastable 2*s* state of hydrogen-like ions [4]. Their method predicts an emission rate:

$$A = 8.2294\, Z^6\ \text{s}^{-1} \tag{1}$$

There is no doubt that such two-photon emission occurs if not pre-empted by some density-sensitive process such as electron collisional interruption of the intermediate state or collisional ionization. The measured rate agrees with Eq. (1): this is nicely shown by measurements of two-photon emission in beam-foil spectroscopy reported by Marrus and Mohr [5]. In the beam-foil experiments, high-velocity hydrogen-like ions from an accelerator are excited when they pass through a thin foil and after they leave the foil, the

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emission ($2s \to 1s$) is measured. Two-photon emission is identified by coincidence detection of the two photons. The angular distribution and energy distribution are measured and agree with theory. In effect, because the ion velocity is accurately known, the excited-state lifetime is measured with a ruler. The measured emission rate agrees with Eq. (1) to within about 1% for H-like ions of Z up to 18. We note that the emission rate rises rapidly with a Z^6 power law.

Multiphoton processes involving visible light from high-intensity lasers have been studied for many years giving rise to a well-developed science of nonlinear optics [6,7]. Recently this science is beginning to advance into the X-ray range [8,9].

3. Two-photon absorption

Two-photon absorption opacity was calculated by More and Rose in 1991 [10,11] using a semi-classical method [12]. The computer code was tested for $2s \to 1s$ emission and agreed with Eq. (1) to within a few percent [10,11]. The opacity process considered was absorption by ground-state H-like Fe at conditions of the solar interior. The absorption was calculated [10,11] by second-order perturbation theory as a sum over intermediate states with dipole matrix elements $R_{n,l}{}^{n',l\pm1}$ linking the initial ($1s$) state to various excited states; the energy denominators ΔE are differences of initial and intermediate state energies, including the photon energies. The calculated absorption cross-section was not large, about 10^{-24} cm^2/ion, comparable to the Compton scattering cross-section. The absorption cross-section is formally independent of Z, however it rises with the eighth power of the principle quantum number n of the electrons involved:

$$\sigma \propto \left| \sum \frac{RR}{\Delta E} \right|^2 \propto n^8 \tag{2}$$

This strong dependence arises because the radial dipole matrix-elements R are proportional to n^2 when the change of n is not large (that scaling already follows from the Bohr model). When the implications of this scaling are appreciated it seems appropriate to re-examine the possibility that two-photon absorption could be important, especially for plasmas in which the ions carry bound electrons with $n > 1$ and where ambient radiation can supply the needed second photon.

The basic theory of two-photon processes is sketched in several textbooks of quantum electrodynamics [13–16]. In this brief paper we describe several difficult points in the calculation of two-photon absorption opacity for plasmas and suggest how these difficulties can be solved.

4. Basic theory

For moderate Z, such as Z = 26, the non-relativistic second-order perturbation theory appears to be sufficiently accurate. The unperturbed Hamiltonian for one electron and the radiation field can be written [13–20]:

$$H_e^0 = \frac{1}{2m} p^2 - eV(r) \tag{3}$$

$$H_{rad}^0 = \sum_{k,s} \hbar\omega_k \left(n_{ks} + \frac{1}{2} \right) \tag{4}$$

The electron-photon coupling is

$$H^{(1)} = -\frac{e}{mc} \vec{p} \cdot \vec{A} \tag{5}$$

A higher-order relativistic term proportional to A^2 is responsible for Compton scattering but can be neglected for present purposes. The radiation field operator is

$$\vec{A}\left(\vec{r}\right) = \sum_{k,s} \hat{e}_{k,s} C_k \left(a_{k,s} e^{i\vec{k}\cdot\vec{r}} + a_{k,s}^{+} e^{-i\vec{k}\cdot\vec{r}} \right) \tag{6}$$

where a^+, a are photon creation and annihilation operators and the normalization C_k is

$$C_k = \left(\frac{2\pi\hbar^2 c^2}{L^3 \hbar\omega} \right)^{1/2} \tag{7}$$

Two-photon processes are predicted by second-order perturbation theory,

$$H_{1\to3}^{(2)} = \sum_{state\ 2} \frac{H_{1\to2}^{(1)} H_{2\to3}^{(1)}}{E_1 - E_2} \tag{8}$$

This leads to a transition rate

$$rate = \sum_{process} \frac{2\pi}{\hbar} \left| H_{1\to3}^{(2)} \right|^2 \rho(E_f) \tag{9}$$

When we expand this expression we can identify the rates for the various two-photon absorption and emission processes, including several Raman effects. Eq. (2) follows from Eqs. (5, 8, 9). In Eq. (9), the word "process" means photon absorption and/or emission, which occur differently for 2-photon emission, 2-photon absorption and for the Raman processes. The symbol $\rho(E_f)$ indicates the appropriate photon density of states, which can be different for the different process.

5. Cross-section and opacity

The first need is to convert Eq. (9) for the rate of two-photon transitions into a cross-section and then average it to calculate the opacity. The experiment observes the attenuation of photons from a hot backlighter source that is a hot CH_2 plasma which has been shocked and compressed by the implosion of a wire array [1]. The backlighter photons are absorbed as they traverse a thin foil target. To find the effective opacity due to two-photon events we sum the cross-sections over all processes able to remove or replace the photons of interest. The second photon might come from the radiation field existing in the plasma, which might be produced by thermal emission in the plasma, or might come from the backlight source. If the second photon is emitted, it might be spontaneous emission or might have been stimulated by existing photons from either source. The opacity summed over the second photon thus depends on the radiation temperature and/or backlight beam-flux. This integrated cross-section has units of cm^2. This is much larger than the tiny cross-section appropriate to nonlinear absorption of two identical photons from an intense X-ray source; that cross-section would have units of cm^4-s.

In this brief paper we do not describe these calculations in detail, because the basic ideas are well known (see especially the two books cited in reference 18).

6. Six processes

A second step is to recognize that there are as many as six processes to consider. Two-photon absorption is most important, but stimulated emission into the detector direction reduces the measured opacity and, depending on the photon energy relative to the difference of atomic energies, there can be two or four Raman effects (Stokes and anti-Stokes). These Raman effects also increase and/or reduce the opacity. Our calculations include all these processes but they are not all equally important. Fig. 1 gives cartoon sketches of the four processes that occur for $h\nu_1 < \Delta E = E_u - E_l$ in a transition from a lower state l to an upper state u.

When $h\nu_1 > \Delta E$, the two-photon emission and absorption cannot occur (for this pair of levels). The Raman processes of Fig. 1 occur, as a continuation of the $h\nu_1 < \Delta E$ Raman processes, but an additional pair of Raman processes also can occur, in which the energy changes