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Optical Pumping of Metastable Helium Atoms in Polarized Electron Sources

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Abstract. — In this paper we present a comparison between the Orsay and Rice University polarized electron sources. Different explanations of the polarization degradation are discussed for the two cases. For the Rice source radiation, trapping can account for the observed reduction in polarization because of the high absorption cross sections associated with the D_1 transition. Some improvements are proposed that might increase the electron polarization.

1. Introduction

Ten years ago Walters *et al.* [1] designed a polarized electron source using a helium afterglow. This was based on the polarization of helium metastable atoms by optical pumping. A chemi-ionization reaction with CO_2 produced free polarized electrons. At that time, this source provided results far superior to those of conventional AsGa sources (polarization limited to 50%). Following a series of improvements the flowing afterglow source could provide a maximum polarization of 87% at electron currents $I_e = 0.1 \mu\text{A}$; but this decreased to 50% for $I_e = 70 \mu\text{A}$ [2].

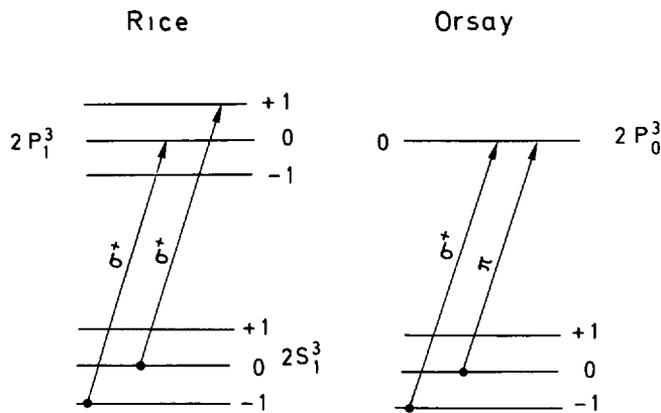
In 1989 the Institut de Physique Nucléaire at Orsay chose the Rice equipment for the projected European Electron Accelerator Facility (ELFE), but with some changes. The results now obtained at Orsay [3] are very close to those obtained with the Rice source and show the same decrease in polarization at high extracted currents. To explain this effect, we studied sometime ago radiation trapping [4] in the Orsay case [5]; we now extend this discussion to the Rice source. More generally we compare the specific characteristics and the performances of the two sources to propose design improvements.

2. Comparison Between the Orsay and Rice Sources

The differences between the two sources [2, 6] are shown in Table I. The main one is the choice at Orsay of the electric dipole D_0 transition (between the $2P_0^3$ and $2S_1^3$ substates) for the optical pumping (Fig. 1). The Doppler width at ambient temperature and the splitting between the $2P_1^3$ and $2P_2^3$ states are 1.7 and 2.4 GHz, respectively. Thus these transitions are partially

Table I. — *Main differences between Rice and Orsay sources.*

	RICE	ORSAY
Gas speed	$\sim 4 \times 10^3 \text{ cm s}^{-1}$	$1.2 \times 10^4 \text{ cm s}^{-1}$
Optical pumping transition	D_1	D_0
Laser	Single mode low power (120 mW)	Multimode, linewidth = 2.5 GHz powerful (1 W)
Extraction	High vacuum 0.200 - 0.50 mb	Fair vacuum 0.05 - 0.10 mb

Fig. 1. — Optical pumping transition used at Rice and Orsay sources with σ^+ and π lights.

overlapping. The Rice group built a single mode, low power (120 mW), stabilized LNA laser [7] and could optically pump on the D_1 transition without exciting the $2P_2^3$ state, which can lead to depolarization. Theoretically the D_1 transition can give 100% polarization with a σ_+ or σ_- light beam. At Orsay, optical pumping is undertaken using a commercial Microcontrol Nd-YAG laser equipped with a LNA crystal [8]. This multimode laser has 2.5 GHz linewidth, but is powerful (1 Watt) and comparatively inexpensive. Pumping on the D_1 transition is very difficult or impossible with this laser without exciting the $J = 2$ state. However with simultaneous σ and π pump beams one can pump using the $2P_0^3$ state which is well separated from the $2P_1^3$ state ($\Delta = 29$ GHz). The effective absorption cross-sections, corrected to take into account the laser width [9], are approximately 6 times higher for the D_1 transition than for the D_0 transition: i.e. the final state density leads to a factor 3, and an additional factor 2 is due to the laser frequency profile.

Another difference between the Rice and Orsay systems is the helium bulk flow velocity in the afterglow, which is a factor of two larger in the Orsay source. Thus the metastable decay lengths [10], and consequently the metastable density and the extracted electron current, are larger at Orsay for the same pressure. But metastable atoms in the $2S_0^1$ state ($\sim 5\%$ concentration depending on the Laval nozzle tuning) also survive passage down the flowtube (the lifetime is longer than the flight time) and can generate unpolarized electrons. At Rice care was exercised to reduce the background gas pressure in the extraction system. This eliminates electrons due to ionization and excitation of the background gas.

3. Rate Equation - Radiation Trapping

The rate equations are different from the Orsay case [5]. If n^+ , n^0 , n^- , N^+ , N^0 , N^- are the densities of $2S_1^3$ and $2P_1^3$ atoms, respectively, in the substates $m = +1, 0, -1$, at a point r and time t , the six equations that describe these densities are:

$$\begin{aligned}
 \frac{dn^-}{dt} &= -\frac{n^-}{\tau_p} + \frac{N^-}{3\tau} + \frac{N^0}{3\tau} - \frac{n^-}{\tau_r} \\
 \frac{dn^0}{dt} &= -\frac{n^0}{\tau_p} + \frac{N^+}{3\tau} + \frac{N^-}{3\tau} - \frac{n^0}{\tau_r} \\
 \frac{dn^+}{dt} &= \frac{N^+}{3\tau} + \frac{N^0}{3\tau} - \frac{n^+}{\tau_r} \\
 \frac{dN^-}{dt} &= -\frac{N^-}{\tau} \\
 \frac{dN^0}{dt} &= \frac{n^-}{\tau_p} - \frac{N^0}{\tau} \\
 \frac{dN^+}{dt} &= \frac{n^0}{\tau_p} - \frac{N^+}{\tau}
 \end{aligned} \tag{1}$$

where τ is the lifetime of the $2P_1^3$ state ($\tau = 10^{-7}$ s) and τ_r is the relaxation time of metastables. The relaxation time τ_r describes phenomenologically the loss of metastables to the walls [11] and is calculated [10, 12] by $\tau_r = \alpha R^2 P / (DP)$ with $DP = 470 \text{ tor.cm}^2\text{s}^{-1}$ and $\alpha = 0.27$, R and P being the radius and the helium pressure in the source, respectively. A complete discussion of the measurement and calculation of τ_r at Rice is presented by Keliher [10]. The quantity τ_p is the so-called pumping time defined by $\frac{1}{\tau_p} = \frac{\sigma_a \cdot I_T}{h\nu_0 \cdot S}$, where I_T is the total laser power distributed over area S ; $h\nu_0$ is the energy quantum for a frequency $\nu_0 = c/\lambda_0$, with $\lambda_0 = 1.083 \mu\text{m}$. We denote by λ_0 the wavelength involved in the transition from the metastable "ground state" $2S_1^3$ to the excited state $2P_1^3$. The level-to-level absorption cross section σ_a is deduced from the classical state-to-state absorption cross section [9] for unpolarized light

$$\bar{\sigma}_a = \frac{g_f}{g_i} \frac{\lambda^2}{8\pi\tau} F(\nu - \nu_0)$$

where

$$F(\nu - \nu_0) = \left[\frac{Mc^2}{2\pi kT\nu_0^2} \right]^{1/2} \exp \left[- \left(\frac{\nu - \nu_0}{\nu_0} \right)^2 \frac{Mc^2}{2kT} \right]$$

is the Doppler distribution around ν_0 [13]; so $\sigma_a = \frac{\lambda^2}{2\pi\tau} F(\nu - \nu_0)$. The laser width correction is not justified in the Rice case because its frequency width is negligible. Note the intensity $I = I_T/S$ is the significant parameter rather than I_T .

Equations (1) describe the metastable populations in a frame following the gas flow. These populations are a function of time only. The deduced metastable polarization

$$Pol = \frac{n^+ - n^-}{n^+ + n^0 + n^-} \quad (2)$$

is not dependent on τ_r , but only on τ_p . This means that the polarization never reaches a steady state ($t \rightarrow \infty$), but if the light is intense enough and/or the pumping area great enough, Pol can be large. This description is slightly different for a sealed cell, where a discharge repopulates the metastable sublevels which are destroyed at the cell wall [14]. In this case, repopulation would introduce additional terms $n/3\tau_r$ in the rate equations. But calculations using the equation system (1) show that the polarization is not very dependent on these terms. To check the validity of the model, we calculated the polarization as a function of laser power, which was measured at Rice [2] for a low metastable density ($n \leq 10^9 \text{ cm}^{-3}$) for which radiation trapping is negligible. Figure 2 demonstrates that the results agree very well with the experimental data.

Radiation trapping is the reabsorption of unpolarized radiation emitted by the decay of excited states during the optical pumping process. Figure 3 shows all the emission and absorption transitions. To take reabsorption into account, we add to the rate equations (1) specific terms following the procedure proposed by Anderson *et al.* [15] For example, the additional term to the third equation in (1) has the shape.

$$-\frac{N^e}{\tau} \int d^3\bar{r}' d\nu [P_{\Delta m}(\theta) F(\nu - \nu_0)] \times \exp[-(\bar{r} - \bar{r}') \sum \sigma_i^k(\theta)(n^a - N^b)]$$

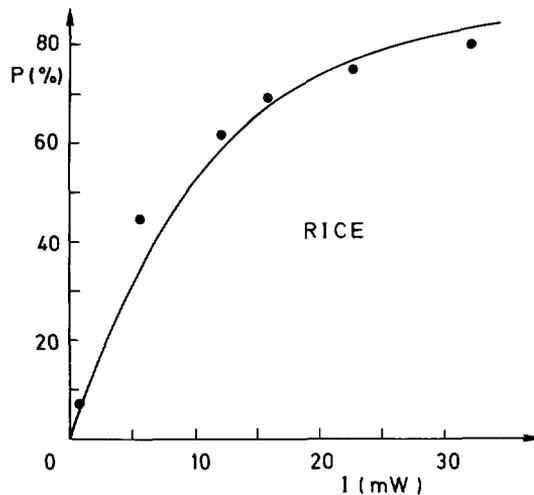


Fig. 2. — Electron polarization as a function of laser power. Experimental data [2] and polarization calculated without radiation trapping by rate equations (1) The parameters are $I_T = 80 \text{ mW}$, $R = 5 \text{ cm}$ and $n = 10^9 \text{ cm}^{-3}$

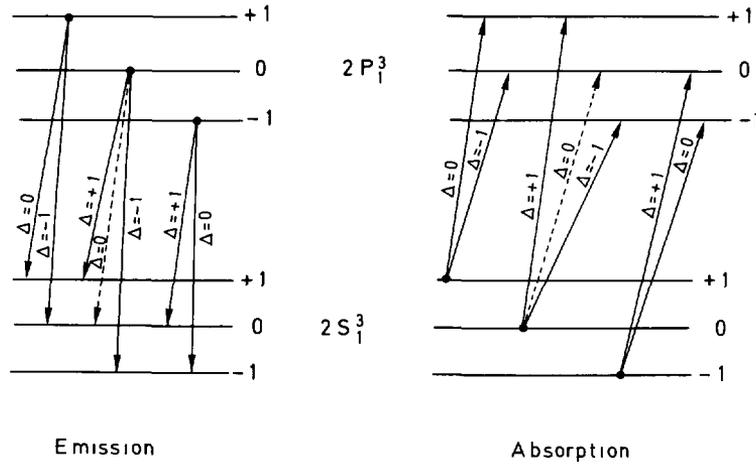


Fig. 3 — Emission and absorption transitions involved in electric dipole transition $2S_1^3 - 2P_1^3$ for unpolarized radiations. $\Delta = m_f - m_i$ is the magnetic quantum number transfer in these transitions. For $\Delta = m_f - m_i = 0$ the transition is forbidden.

$$\times \left[\sum \sigma_l^k(\theta)(n^0 - N^d) \right] \tag{3}$$

where θ is the angle between $\bar{r} - \bar{r}'$ and the direction of the magnetic field.

The first factor is the electric dipole deexcitation probability of the N^e atoms in $2P_1$ state at the point \bar{r}' . This radiation is attenuated along the path $\bar{r} - \bar{r}'$ by the transition $a \rightarrow b$ (second factor). The last factor gives the radiation absorption by the transition $c \rightarrow d$ at the point \bar{r} . The absorption cross section σ_l^k corresponds to the absorption of emitted radiation characterised by the transition $\Delta m = (m_f - m_i) = k$ and absorbed by the transition $\Delta m = (m_f - m_i) = l$. These quantities are deduced from the matrix elements of the dipole operator \bar{D} as $|\langle i|\bar{E}_\alpha(\theta)\bar{D}|f\rangle|^2$, $\bar{E}_\alpha(\theta)$ being the field of the incident wave [7]. Appendix gives the nine values of $\sigma_l^k(\theta)$ (available only for the D_1 transition in helium pumping) and the additional term (3) after simplification (same example of the third Eq. (1)).

The NAG code from the CERN library is used to solve the equation system (1 + 3). The input parameters are:

- a) laser power $I_T = 80$ mW
- b) irradiation area $S = 40$ cm².

4. Results and Discussion

The rate equations give the metastable densities as a function of time. We have calculated the metastable polarization using a flight time corresponding to a length of approximately 6 cm. Note that the experimental uncertainties ($\lambda/4$ plate, optics, chemi-ionization efficiency, spin singlet $2S_0^1$ metastable presence, etc.) are not included although they limit the attainable polarization to below 100%. These corrections are estimated to be approximately 10% [2]. As the maximum polarization at Rice is 87%, this value is chosen as the normalization factor of the calculated polarization.

The most serious assumption of the previous calculation (Eq. (4)) was that n^i and N^j densities are independent of the position \bar{r} whereas they have an approximately parabolic radial profile [12,17]. An improvement to the calculations previously presented is to assume that the densities have a $\cos(\pi r/2R)$ spatial distribution. This assumption is more realistic because the radiation emitted at $r = R$ has a higher probability to escape than at the center ($r = 0$). Unfortunately, by including density variation, the computer times become unreasonably long. Certainly a more sophisticated procedure must be developed, as suggested by Happer and Redsun [18]. Nevertheless, for a rough understanding of the experimental data of the Rice group the previous calculations are sufficient. In the future we hope to obtain more accurate metastable polarization data and to improve the comparison with sophisticated calculations.

In the present work the cosine function distribution is used only to calculate the polarization values versus the metastable density (Fig. 4) because the results with and without the density correction are very close: at high density ($n > 10^{10} \text{ cm}^{-3}$) this correction increases the polarization by 10% approximately. Experimental data previously published [2] are shown on the same figure. The corresponding metastable concentrations were determined by absorption of the light emitted by a helium lamp [10] and are inaccurate. Nevertheless, one sees an agreement despite the crudeness of the reabsorption model. The conclusion is that radiation trapping is responsible for the polarization decrease at high metastable concentrations or high electron currents. Fortunately the Rice results can be improved by changing the experimental set-up and procedure: laser, geometry, etc. Figure 4 also shows the metastable polarization for the Orsay source. In this case the trapping becomes significant only at metastable densities higher than those used at Rice because the effective absorption cross-section is lower for the D_0 transition than for the D_1 transition. This proves the value of optical pumping using the D_0 transition.

The polarization is calculated as a function of the total laser power (Fig. 5) for two metastable densities (7×10^9 and 10^{10} cm^{-3}). This calculation shows that saturation is almost reached at $I_T = 80 \text{ mW}$, as was confirmed by the Rice measurements. Increasing the laser power improves

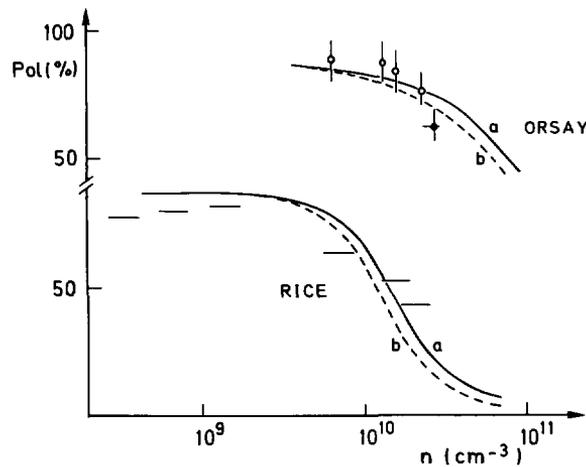


Fig. 4. — Polarization calculated *versus* the metastable density for the Orsay and Rice sources. The cosine profile is included (a) or not (b). The experimental data are presented with error bars. The empty points correspond to the metastable density measured by laser at Orsay. The other ones (full point and lines) correspond to densities measured by helium light absorption.

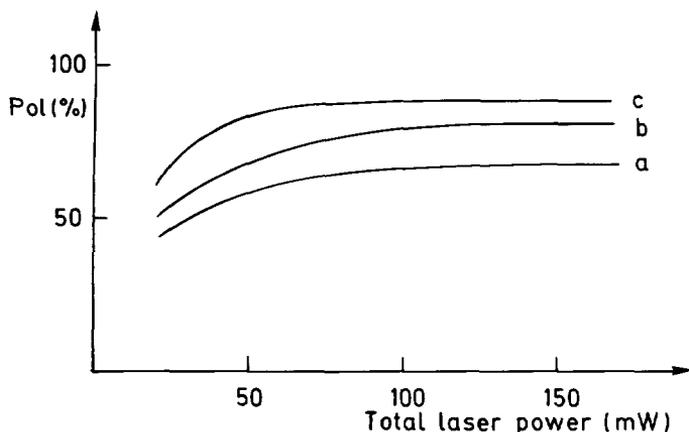


Fig. 5. — Polarization calculated versus the total laser power for metastable density of 10^{10} cm^{-3} (a) and $7 \times 10^9 \text{ cm}^{-3}$ (b). The curve (c) is the polarization calculated without radiation trapping correction.

the polarization only slightly. We have assumed that the laser light beam is homogeneous over the pumping volume. In fact, measurements of the light distribution at point r show a Gaussian distribution. This could lead to the unsaturation of some metastables. This effect must be negligible with the Rice set-up, where two baffles concentrate the metastable beam and increase the polarization by 10% [19]. On the other hand it must play a large role at Orsay. Figure 5 also shows the polarization *versus* the laser power, calculated without the trapping corrections. Then the polarization is 87% except at low power (I_T less than 50 mW) when the pumping time τ_p becomes large.

To reduce the radiation trapping it seems interesting to reduce the dimension of the optical pumping chamber. In this way the metastable density, i.e. the electron current, decreases (the decay length is shorter [10, 12]). Therefore an optimization process is required.

5. Conclusion

From our calculations, it appears that the polarization degradation of the Rice source is mostly due to radiation trapping, as it was suspected by Walters *et al.* [20,21]. The curves in Figure 4 show clearly that this effect is more important at Rice than at Orsay because the absorption cross-sections are higher for the D_1 transition: the factor 6 between these cross sections explains approximately the shift between the curves. Since spin polarization is conserved in chemi-ionization reactions (checked at Rice) and the extraction geometry is identical in the two sources, one expects electron polarization superior at Orsay than at Rice for the same high electron currents. In fact, the experimental values of these polarizations [2, 3] are close. This disagreement is presently studied [22].

To increase the polarization at Rice it would be fruitful to use optical pumping by D_0 transition under the present conditions. After that, some minor improvements in the experimental conditions could be tested to increase the polarization: i) an increase of the laser power per unit area, ii) an improvement of the electron extraction efficiency so as to work at lower metastable densities: in this way, a higher polarization should be reached for the same current, and iii) a reduction of the pumping volume to reduce the trapping probability. Finally, one could use a

classical technique, i.e. decrease the effective absorption cross sections by detuning the laser frequency to the wings of the atomic resonance line [23].

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Appendix

The same assumptions as those previously proposed by Anderson *et al.* [15] are used to simplify equations (3):

- i) the pumping chamber is a infinitely long cylinder of radius R ($R = 5$ cm) with non-reflective walls,
- ii) the densities n^+ and N^+ are taken to be independent of position \bar{r} ,
- iii) the laser light is homogeneous. The integrations over frequency and angle (Eqs. (3)) are performed using Hermitian and Gaussian integration procedures. The integration over radius is performed exactly.

The trapping contribution has then the simplest shape $T_1 + T_2 + T_3$ where:

$$T_1 = \frac{(N^0 + N^-)}{2\sqrt{\pi\tau}} \sum_i \omega_i \sum_j \omega_{3j} F_3(u_j)(n^+ - N^+) + \omega_{1j} F_1(u_j)(n^+ - N^0)$$

with

$$F_3(u_j) = 2\sqrt{1 - u_j^2} \left[\frac{1 - \exp\left(-\frac{9\gamma_i R \phi(u_j)}{4u_j}\right)}{\phi(u_j)} \right]$$

and

$$F_1(u_j) = \frac{(2 - u_j^2)^2}{\sqrt{1 - u_j^2}} \left[\frac{1 - \exp\left(-\frac{9\gamma_i R \phi(u_j)}{4u_j}\right)}{\phi(u_j)} \right]$$

and

$$\begin{aligned} \phi(u_j) = & \frac{u_j^4}{(2 - u_j^2)} (n^0 + n^- - N^+ - N^0) + (2 - u_j^2)(n^+ + n^0 - N^-) + 2 \frac{(1 - u_j^2)}{(2 - u_j^2)} u_j^2 \\ & \times (n^+ + n^0 + n^- - N^+ - N^0 - N^-) \end{aligned}$$

$$T_2 = \frac{(N^+ + N^0 + N^-)}{\sqrt{\pi}\tau} \sum_i \omega_i \sum_j \omega_{5j} (n^+ - N^+) F_5(u_j) + \omega_{3j} (n^+ - N^0) F_3(u_j) + \omega_{3j} (n^+ - N^0) F_3(u_j) \quad (4)$$

with

$$F_5(u_j) = \frac{2}{\sqrt{1-u_j^2}} \left[\frac{1 - \exp\left(-\frac{9\gamma_i R\phi(u_j)}{4u_j}\right)}{\phi(u_j)} \right]$$

and

$$F_3(u_j) = \sqrt{1-u_j^2} \left[\frac{1 - \exp\left(-\frac{9\gamma_i R\phi(u_j)}{4u_j}\right)}{\phi(u_j)} \right]$$

and

$$\phi(u_j) = (1-u_j^2)(n^+ + 2n^0 + n^- - N^+ - 2N^0 - N^-) + 2u_j^2(n^+ + n^0 + n^- - N^+ - N^0 - N^-)$$

$$T_3 = \frac{(N^+ + N^0)}{2\sqrt{\pi}\tau} \sum_i \omega \sum_j \omega_{3j} F_3(u_j)(n^+ - N^+) + \omega_{5j} F_5(u_j)(n^+ - n^0) \quad (5)$$

with

$$F_3(u_j) = 2\sqrt{1-u_j^2} \left[\frac{1 - \exp\left(-\frac{9\gamma_i R\phi(u_j)}{4u_j}\right)}{\phi(u_j)} \right]$$

and

$$F_5(u_j) = \frac{1}{\sqrt{1-u_j^2}} \left[\frac{1 - \exp\left(-\frac{9\gamma_i R\phi(u_j)}{4u_j}\right)}{\phi(u_j)} \right]$$

and

$$\begin{aligned} \phi(u_j) &= (2-u_j^2)(n^0 + n^- - N^+ - N^0) + \frac{U_j^4}{(2-u_j^2)}(n^+ + n^0 - N^0 - N^-) \\ &+ \frac{2u_j^2(1-u_j^2)}{2-u_j^2}(n^+ + n^0 + n^- - N^+ - N^0 - N^-) \end{aligned}$$

χ_i are the nodes and ω_i the weighting factors of the Hermite integration method [24], and u_j the nodes and ω_{ij} the weighting factors of the Gaussian integration method [24], with:

$$\gamma_i = \frac{\lambda^3}{8\pi\tau} \left[\frac{M}{2\pi kT} \right]^{1/2} e^{-\chi_i^2}.$$

The cross-sections $\sigma_{\Delta m, \text{absorption}}^{\Delta m, \text{emission}}$ are

$$\begin{aligned}\sigma_0^0 &= 2 \sin^2 \theta \cdot \sigma \\ \sigma_+^0 &= \sigma_-^0 = \cos^2 \theta \cdot \sigma \\ \sigma_+^+ &= \sigma_-^- = \frac{\sin^4 \theta}{(1 + \cos^2 \theta)} \cdot \sigma \\ \sigma_0^+ &= \sigma_0^- = \frac{2 \cos^2 \theta \cdot \sin^2 \theta}{(1 + \cos^2 \theta)} \cdot \sigma \\ \sigma_-^+ &= \sigma_+^- = (1 + \cos^2 \theta) \cdot \sigma \\ \sigma &= \frac{3\lambda^2}{32\pi\tau} F(\nu - \nu_0)\end{aligned}$$

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