Free-free radiative absorption coefficient for the negative argon ion*

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The free-free absorption coefficient for Ar^- is calculated at radiation wavelengths of 2000 and 20000 Å for various temperatures. Electron correlation corrections to the Hartree-Fock approximation are obtained by using many-body perturbation theory. Comparison is made with other theoretical methods and experiment.

I. INTRODUCTION

The free-free radiative absorption process¹ has long been of interest in determining both opacities in $stars^2$ and the continuous spectra of atomic gases.³ Free-free absorption is also a mechanism by which the plasma absorbs energy in the laserdriven fusion process.⁴ In recent years, accurate values for the H⁻ free-free absorption coefficient have been calculated^{5,6} using polarized-orbital theory.⁷ Semiempirical methods^{1,8} have been used with considerable success in determining free-free coefficients for more complex atoms. At the very long wavelengths asymptotic methods^{9,10} have been shown^{11,12} to be fairly accurate in computing freefree coefficients. In this paper we apply manybody perturbation theory to an *ab initio* calculation of the free-free absorption coefficient of Ar⁻. The influence of the radiation wavelength on the magnitude of the Coulomb correlation corrections to the Hartree-Fock approximation is determined. The final results are compared with other theoretical methods and experiment. In Sec. II many-body perturbation theory is reviewed and its application to free-free scattering coefficients is discussed. The numerical results and our conclusions are given in Sec. III.

II. THEORY

The free-free absorption cross section for a negative ion may be calculated using the many-body perturbation theory of Brueckner¹³ and Goldstone.¹⁴ The Hamiltonian H of the free electron and the N-electron neutral atom is given by

$$H = H_{0A} + H_{corr} , \qquad (1)$$

where

$$H_{0A} = \sum_{j=1}^{N+1} \left(-\frac{\hbar^2}{2m} \nabla_j^2 - \frac{Ze^2}{r_j} + V_j \right), \qquad (2)$$

$$H_{\text{corr}} = \sum_{i < j=1}^{N+1} v_{ij} - \sum_{j=1}^{N+1} V_j .$$
(3)

The single-particle potential V_j is generally the Hartree-Fock potential, while v_{ij} is the Coulomb interaction between electrons *i* and *j*. In Eq. (2), *m* is the electron mass, *e* is the magnitude of the electronic charge, and *Z* is the atomic number. The *N* lowest energy single-particle solutions ϕ_n , with eigenenergies ϵ_n , of

$$\left(-\frac{\hbar^2}{2m}\nabla^2 - \frac{Ze^2}{r} + V\right)\phi_n = \epsilon_n\phi_n , \qquad (4)$$

form the unperturbed Slater determinantal ground state Φ_0^N of the atom. A positive-energy solution ϕ_k of Eq. (4), with momentum $\hbar k$, can be used to form the determinantal function (ϕ_k, Φ_0^N) for the electron-atom system.

The unperturbed Hamiltonian H_0 for the timedependent perturbation expansion is given by

$$H_0 = H_{0A} + H_{rad},$$
 (5)

where H_{rad} is the Hamiltonian for a quantized electromagnetic field.¹⁵ The eigenstates $\tilde{\Phi}_{j}^{n}$ of H_{0} are products of a state $(\phi_{k_{j}}\Phi_{0}^{N})$ and a radiation state with *n* photons of frequency ω or wavelength λ . The eigenenergy $E_{j}^{n} = E_{0}^{N} + \hbar^{2}k_{j}^{2}/2m + n\hbar \omega$, where E_{0}^{N} is the ground-state energy associated with the state Φ_{0}^{N} . The perturbation Hamiltonian H_{1} is then

$$H_1 = H_{\rm corr} + H_{\rm int} , \qquad (6)$$

where

$$H_{\text{int}} = \sum_{j=1}^{N+1} \left(-\frac{ie\,\hbar}{mc} \vec{\mathbf{A}}(\vec{\mathbf{r}}_j) \cdot \vec{\nabla}_j + \frac{e^2}{2mc^2} \left| \vec{\mathbf{A}}(\vec{\mathbf{r}}_j) \right|^2 \right), \quad (7)$$

and $\vec{A}(\vec{r})$ is the quantized vector potential.¹⁵ The solution $\Psi(t)$ of the time-dependent Schrödinger equation with Hamiltonian $H = H_0 + H_1$ is given by¹⁶

$$\Psi(t) = \lim_{\gamma \to 0} \frac{e^{-iH_0 t/\hbar} U_{\gamma}(t) |\tilde{\Phi}_i^n\rangle}{\langle \tilde{\Phi}_i^n | U_{\gamma}(t) | \tilde{\Phi}_i^n\rangle},$$
(8)

where $|\tilde{\Phi}_i^n\rangle$ is the initial state. The time-evolution operator¹⁷ $U_{\gamma}(t)$ is given as

$$U_{\gamma}(t) = 1 + \frac{1}{i\hbar} \int_{-\infty}^{t} d\tau H_{1}(\tau) + \frac{1}{(i\hbar)^{2}} \int_{-\infty}^{t} d\tau \int_{-\infty}^{t} d\tau' H_{1}(\tau) H_{1}(\tau') + \cdots, \quad (9)$$

where

$$H_1(t) = e^{iH_0t/\hbar} H_1 e^{\gamma t} e^{-iH_0t/\hbar} , \qquad (10)$$

and γ is a small positive number which causes $H_1(t)$ to be "turned on" adiabatically. Carrying out the time integrations in Eq. (9), the transition rate w from the state $|\tilde{\Phi}_i^n\rangle$ to the final state $|\tilde{\Phi}_j^{n-1}\rangle$ with the absorption of a photon is given by¹⁸

$$w = \frac{2\pi}{\hbar} \delta(E_i^n - E_f^{n-1})$$

$$\times \left| \langle \tilde{\Phi}_f^{n-1} | \sum_L H_1 \dots \frac{1}{E_i^n - H_0 + i\eta} H_1 \left| \tilde{\Phi}_i^n \rangle \right|^2 \langle \Psi | \Psi \rangle^{-1}, \qquad (11)$$

where \sum_{L} means that only linked Goldstone diagrams^{13,14} are to be included and the term $+i\eta$ ensures the proper boundary conditions. For photoabsorption processes in argon, the normalization factor $\langle \Psi | \Psi \rangle$ was found¹⁹ to be very close to unity.

Techniques to evaluate diagrams developed previously²⁰ are used to evaluate Eq. (11). Diagrams for several different orders of the perturbation expansion are shown in Fig. 1. The solid dots indicate an interaction with the radiation field while the dashed lines not ending in dots represent Coulomb correlation interactions. Figure 1(a) is the lowest-order term yielding the Hartree-Fock result when V of Eq. (4) is chosen to be the Hartree-Fock potential. Diagrams of first order in the Coulomb correlations are shown in Figs. 1(b) and 1(c). There are also exchange diagrams associated with Figs. 1(b)and 1(c), but they are not shown. The second-order direct diagrams in Figs. 1(d) and 1(e) along with their associated exchanges may make significant contributions to the lowest-order free-free cross section. There are many other second-order diagrams, but we expect²¹ that 1(d) and 1(e) are the most important at long wavelengths. These second-order polarization diagrams are difficult to evaluate directly, so that various approximate methods are used to represent their effect.

The lowest-order cross section $d\sigma$, obtained from Eq. (11) in the length formulation of the dipole approximation, is given by

$$d\sigma = \frac{4\pi^2 e^2 \omega}{c} \left| \left\langle \phi_{kf} \right| \epsilon \cdot \vec{r} \left| \phi_{ki} \right\rangle \right|^2 \delta(\epsilon_i - \epsilon_f + \hbar \omega) , \quad (12)$$

where $\epsilon_i = \hbar^2 k_i^2/2m$. In order to evaluate¹ the matrix element of Eq. (12), one may make a partial-wave expansion of ϕ_k to yield

$$\phi_{k} = \frac{1}{kr} \sum_{l=0}^{\infty} i^{l} e^{i\delta_{l}} (2l+1) u_{kl}(r) P_{l}(\cos\theta) , \qquad (13)$$

where the continuum is normalized according to

$$u_{kl}(r) \xrightarrow{r} kr \left[\cos \delta_l j_l(kr) - \sin \delta_l \eta_l(kr) \right], \qquad (14)$$

 $j_{l}(kr)$ and $\eta_{l}(kr)$ are spherical Bessel functions, and δ_{l} is the phase shift. The free-free cross section $\sigma^{FF}(k_{i}, \omega)$ is obtained¹ in lowest order by summing $d\sigma$ of Eq. (12) over all final electron states, averaging over initial directions \hat{k}_{i} , and dividing by the density of scattering centers. The result in units of cm⁵ is given by

$$\sigma_{L}^{\text{FF}}(k_{i},\omega) = 32\pi^{2} \alpha \frac{m}{\hbar} \frac{\omega}{k_{f}k_{i}^{2}} \times \sum_{l=0}^{\infty} \frac{l+1}{3} \left(\left| M_{l+1,l}^{L} \right|^{2} + \left| M_{l,l+1}^{L} \right|^{2} \right),$$
(15)



FIG. 1. Many-body diagrams which contribute to the free-free absorption cross section. Solid dots indicate an interaction with the radiation field, while the dashed lines not ending in dots represent Coulomb interactions. (a) is the lowest-order term and gives the Hartree-Fock result when the single-particle states, labeled k_0 and k_1 , are calculated using the Hartree-Fock potential. (b) and (c) are first-order ground-state and final-state correlation diagrams respectively. Diagrams (d) and (e) are important second-order direct polarization diagrams. There are exchange diagrams not shown that are associated with diagrams (b)-(e).

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where

$$M_{lfli}^{L} = \int_{0}^{\infty} u_{kfli}(r) u_{klli}(r) r \, dr \,, \tag{16}$$

and $k_f = (k_i^2 + 2m \omega/\hbar)^{1/2}$. The velocity form of the free-free cross section $\sigma_V^{FF}(k_i, \omega)$ is given by

$$\sigma_{V}^{FF}(k_{i},\omega) = 32\pi^{2} \alpha \frac{\hbar}{m} \frac{1}{\omega k_{f} k_{i}^{2}} \times \sum_{l=0}^{\infty} \frac{l+1}{3} \left(\left| M_{l+1,l}^{V} \right|^{2} + \left| M_{l,l+1}^{V} \right|^{2} \right),$$
(17)

where

$$M_{lfli}^{V} = \int_{0}^{\infty} u_{kflf}(r) \left(\frac{d}{dr} + \frac{l_{i}(l_{i}+1) - l_{f}(l_{f}+1)}{2r} \right) u_{kill}(r) dr ,$$
(18)

and α is the fine-structure constant. Higher-order diagrams, when separated into their various angular momentum components,²⁰ add to the terms inside the absolute squares of Eqs. (15) and (17).

The free-free absorption coefficient κ (in cm⁵) is given by¹



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where

$$f(k_{i}) = 4\pi \left(\frac{\hbar^{2}}{2\pi m k_{B}T}\right)^{3/2} k_{i}^{2} \exp\left(-\frac{\hbar^{2} k_{i}^{2}}{2m k_{B}T}\right)$$
(20)

is the Maxwell-Boltzmann distribution of electrons per unit volume, normalized to unity.²² In Eq. (20) k_B is Boltzmann's constant and T is the temperature in degrees Kelvin.

III. RESULTS AND DISCUSSION

The single-particle scattering states needed for the evaluation of the lowest-order Ar⁻ free-free absorption cross section were calculated numerically²⁰ as fixed core Hartree-Fock $3p^{6} {}^{1}Skl^{2}L$ continuum wave functions. The radial matrix elements of Eqs. (16) and (18) were obtained by adding a numerical integration part (0 - R) to an asymptotic part $(R \rightarrow \infty)$ calculated analytically by making the integral absolutely convergent.⁵ The asymptotic form for $u_{kl}(r)$ is given by Eq. (14). For example, the asymptotic part of the $k_0 s - k_1 p$ matrix element of Eq. (16) is given by





FIG. 2. Hartree-Fock free-free absorption partialwave cross sections for Ar⁻ at $\lambda = 2000$ Å (in units of cm⁵). The length form of the dipole operator is used. (a) $s \rightarrow p$, (b) $p \rightarrow s$, (c) $p \rightarrow d$, (d) $d \rightarrow p$, (e) $d \rightarrow f$, (f) $f \rightarrow d$, (g) total cross section $(s \rightarrow p, p \rightarrow d, d \rightarrow f)$.

FIG. 3. Hartree-Fock free-free absorption partialwave cross sections for Ar⁻ at $\lambda = 20000$ Å (in units of cm⁵). The length form of the dipole operator is used. (a) $s \rightarrow p$, (b) $p \rightarrow s$, (c) $p \rightarrow d$, (d) $d \rightarrow p$, (e) $d \rightarrow f$, (f) $f \rightarrow d$, (g) total cross section $(s \rightarrow p, p \rightarrow d, d \rightarrow f)$.

$$\int_{R}^{\infty} u_{k_{1}p}(r) u_{k_{0}s}(r) r \, dr = \lim_{\epsilon \to 0} \int_{R}^{\infty} e^{-\epsilon r} \left(\frac{\sin(k_{1}r + \delta_{p})}{k_{1}r} - \cos(k_{1}r + \delta_{p}) \right) \sin(k_{0}r + \delta_{s}) r \, dr$$

$$= \frac{1}{2} \left(\frac{\sin z^{*}}{(\Delta k^{*})^{2}} - \frac{\sin z^{-}}{(\Delta k^{*})^{2}} - \frac{R\cos z^{*}}{\Delta k^{*}} + \frac{R\cos z^{-}}{\Delta k^{*}} + \frac{\sin z^{*}}{k_{1}\Delta k^{*}} - \frac{\sin z^{-}}{k_{1}\Delta k^{*}} \right), \tag{21}$$

where $z^* = \Delta k^* R + \Delta \delta^*$, $\Delta k^* = k_1 \pm k_0$, and $\Delta \delta^* = \delta_p \pm \delta_s$. The analytic asymptotic parts for the transitions $s \rightarrow p$, $p \rightarrow d$, and $d \rightarrow f$ were evaluated in the length form, while the $s \rightarrow p$ and $p \rightarrow d$ transitions were also found in the velocity form. By varying the cutoff R from $40a_0$ to $80a_0$ (a_0 is the Bohr radius), it was found that the procedure used above for computing radial matrix elements is accurate to better than 10%.

Hartree-Fock partial-wave free-free absorption cross sections for Ar⁻, computed in the length form using Eq. (15), are shown in Fig. 2 at λ = 2000 Å and in Fig. 3 at λ = 20 000 Å. In computing the free-free absorption coefficient by Eq. (19), it should be noted that the mean value of the Maxwell-Boltzmann momentum distribution, given by Eq. (20), ranges from \overline{k} = 0.090 a_0^{-1} for T = 1000 °K to \overline{k} = 0.284 a_0^{-1} for T = 10000 °K. It was thus found



FIG. 4. Free-free total absorption cross section for Ar⁻ at $\lambda = 2000$ Å (total includes $s \leftrightarrow p$, $p \leftrightarrow d$ in units of cm⁵). (a) Hartree-Fock, length form. (b) Hartree-Fock, volocity form. (c) Hartree-Fock with first-order correlations, length form. (d) Hartree-Fock with firstorder correlations, velocity form. (e) Polarization potential orbitals with first-order correlations, length form. (f) Polarization-potential orbitals with firstorder correlations, velocity form.

sufficient to limit the integration in Eq. (19) to the range k = 0.0 to $1.0a_0^{-1}$. Furthermore the d - f cross sections were found to contribute only 2% to the total Hartree-Fock absorption coefficient in the length form at T = 10000 °K, while being completely negligible at the lower temperatures. Subsequently, as shown in Figs. 4 and 5 where the Hartree-Fock length and velocity total cross sections are compared with later correlated results, only the s - p and p - d partial waves are included in the total expansion. The Hartree-Fock free-free absorption coefficients for Ar⁻, at several temperatures in both the length and velocity forms, are given in Tables I and II.

The first-order electron correlation corrections to the Hartree-Fock free-free cross section were computed by evaluating diagrams (b) and (c) of Fig. 1 and their corresponding exchange terms.



FIG. 5. Free-free total absorption cross section for Ar⁻ at $\lambda = 20000$ Å (total includes $s \leftrightarrow p$, $p \leftrightarrow d$ in units of cm⁵). (a) Hartree-Fock, length form. (b) Hartree-Fock, velocity form. (c) Hartree-Fock with first-order correlations, length form. (d) Hartree-Fock with firstorder correlations, velocity form. (e) Polarizationpotential orbitals with first-order correlations, length form. (f) Polarization-potential orbitals with firstorder correlations, velocity form.

	<i>T</i> = 1000 °K	2000 °K	5000 °K	10 000 °K
Hartree-Fock length	0.16	0.27	0.56	1.03
Hartree-Fock velocity	0.18	0.26	0.48	0.84
HF with first-order ^a length	0.55	0.68	1.07	1.67
HF with first-order ^a velocity	0.44	0.56	0.87	1.37
Polarization potential with first-order length ^a	0.59	0.84	1.49	2.45
Polarization potential with first-order velocity ^a	0.48	0.67	1.19	1.97
Semiempirical ^b (extrapolated)		0.4	0.8	1.5

TABLE I. Free-free absorption coefficients for Ar⁻ at $\lambda = 2000$ Å (in units of 10^{-40} cm⁵).

^a First-order correlation corrections included.

^b See Ref. 8.

The argon l=2 atomic states were calculated numerically as fixed core Hartree-Fock $3p^{52}Pkd^{1}P$ bound and continuum wave functions. Only the important 3p - nd, $kd^{1}P$ atomic excitations were included in the diagram evaluation since previous photoionization work¹⁹ has shown that the 3p-ns, ks¹P excitations contribute only 10%, while inner-shell contributions are even smaller when the length and velocity forms are used.²³ Correlation corrections to the s - p and p - d partial cross sections were made and the summed results are shown in Fig. 4 for $\lambda = 2000$ Å and Fig. 5 for $\lambda = 20000$ Å. Comparing Figs. 4 and 5, the firstorder correlations are seen to make a larger correction to the Hartree-Fock result at the shorter wavelength. In fact at $\lambda = 1067$ Å,²⁴ the first resonance in the free-free cross section occurs when the denominator of Fig. 1(c) and its exchange vanish for the 3p - 4s atomic excitation. At still shorter wavelengths, resonances due to 3p - nsand 3p - nd excitations will occur. The free-free absorption coefficients obtained by adding first-order correlations to the lowest-order Hartree-Fock results are given in Tables I and II.

The important second-order direct polarization diagrams of Fig. 1(d) and (e) were accounted for,²⁵ in an approximate manner, by adding an adiabatic polarization potential to a Hartree-Fock potential and solving Eq. (4) for new scattering orbitals. The polarization potential $V_{pol}(r)$ was numerically calculated using perturbation theory.²⁰ Only the dipole 3p - nd, $kd^{1}P$ excitations were used to evaluate the lowest-order expression for the polarization potential of argon, given by

$$V_{\text{pol}}^{\text{Ar}}(r) = \frac{4}{3}e^4 \sum_{\mu} \frac{|\int_0^\infty u_{kd}(r')(r_{\leq}/r_{>}^2)u_{3\mu}(r')\,dr'|^2}{\epsilon_{3\mu} - \epsilon_{kd}},$$
(22)

where $r_{\zeta}/r_{2}^{2} = r'/r^{2}$ for r' < r and $r_{\zeta}/r_{\zeta}^{2} = r/(r')^{2}$ for r' > r, \sum_{k} represents a sum over bound states and integration over continuum states, the factor $\frac{4}{3}$ comes from angular integrations, and ϵ_{k} are single-particle energies. Since as $r \rightarrow \infty$, $V_{pol}(r) \rightarrow -\alpha_{s}e^{2}/2r^{4}$, Eq. (22) yields a polarizability of

	$T = 1000 \ ^{\circ}\text{K}$	2000 °K	5000 °K	9900 °K	10 000 °K
Hartree-Fock length	0.17	0.24	0.50	1.14	1.16
Hartree-Fock velocity	0.18	0.25	0.50	1.14	1.15
HF with first-order ^a length	0.21	0.28	0.56	1.25	1.27
HF with first-order ^a velocity	0.20	0.27	0.54	1.21	1.23
Polarization potential with first-order ^a length	0.006	0.023	0.14	0.66	0.68
Polarization potential with first-order ^a velocity	0.005	0.022	0.14	0.64	0.65
Semiempirical ^b Asymptotic method ^c velocity Experiment ^d	0.013	0.041	0.22	0.84 0.72 3.8	0.85

TABLE II. Free-free absorption coefficients for Ar⁻ at $\lambda = 20000$ Å (in units of 10^{-38} cm⁵).

^a First-order correlation corrections included.

^b See Ref. 8.

^c See Ref. 29.

^dSee Ref. 30.

Partial wave	k ^a	Hartree-Fock potential	Polarization potential	Optical potential ^b	Polarized orbitals ^c
<i>l</i> = 0 0.2 0.5 0.7 1.0	0.25	9.05	9.34	9.30	9.28
	0.5	8.65	8.94	8.90	8.91
	0.75	8.25	8.51	8.49	8.49
	1.0	7.88	8.11	8.11	8.07
<i>l</i> = 1	0.25	6.23	6.31	6.29	6.31
	0.5	6.00	6.18	6.13	6.16
	0.75	5.70	5.92	5.87	5.89
	1.0	5.41	5.64	6.56	5.60
<i>l</i> = 2	0.25	0.00	0.02	•••	0.03
	0.5	0.05	0.16	0.12	0.15
	0.75	0.28	0.80	0.44	0.65
	1.0	0.86	1.65	1.10	1.43

TABLE III. Phase shifts (in rad.) for scattering orbitals.

^ak is the magnitude of the incident electron momentum (in a_0^{-1}).

^b See Ref. 27.

^c See Ref. 28.

 $\alpha_{d} = 10.26 a_{0}^{3}$. The experimental value for argon is $\alpha_{d} = 11.0a_{0}^{3.26}$ Monopole terms were omitted since they tend to be cancelled²⁵ by those short-range exchange effects corresponding to the exchange diagrams of Fig. 1(d) and (e). Of course, the more difficult direct evaluation of the second-order diagrams would include exchange, polarization, and dynamic effects in a systematic manner. The l=0and l=1 phase shifts for the new orbitals calculated using the polarization potential of Eq. (22) are in good agreement with previous optical-potential²⁷ and polarized-orbital calculations²⁸ for argon as shown in Table III. The difference between the l=2 polarization-potential phase shifts and the optical-potential results are probably due to dynamical effects. We expect from previous investigations of second-order diagrams^{21,25} that 1(d) and 1(e) and their exchanges are the most important at the long wavelengths involved in freefree scattering.

Polarization-potential scattering orbitals were used to evaluate the $s \leftrightarrow p$ and $p \leftrightarrow d$ free-free partial cross sections. The results for the total cross section, including first-order correlations and second-order polarization effects, are shown in Figs. 4 and 5. The second-order corrections are quite large at $\lambda = 20\,000$ Å while their effect at $\lambda = 2000$ Å is not as dramatic. The free-free absorption coefficients for Ar⁻, including firstand second-order correlations, are given in Tables I and II. The semiempirical results of Geltman⁸ at $\lambda = 20\,000$ Å and his results extrapolated to $\lambda = 2000$ Å are given in Tables I and II. Also given at $\lambda = 20000$ Å and T = 9900 °K is an asymptotic method result of John and Williams.²⁹ The asymptotic method consists of setting R = 0 in expressions like Eq. (21) and evaluating the cross sections from the resulting matrix elements. The three different theoretical results agree somewhat, but are in marked contrast with the experimental point of Taylor and Caledonia.³⁰ In fact, inclusion of the stimulated emission factor, $[1 - \exp(-\hbar \omega/k_BT)]$, in Eq. (19), will lower the three theoretical values; in the case of our work from $\kappa(\omega, T) = 6.6 \times 10^{-39}$ cm⁵ to $\kappa(\omega, T) = 3.4 \times 10^{-39}$ cm⁵.

It can be concluded that theoretical free-free absorption coefficients for negative ions are quite difficult to determine accurately. Near the photoionization threshold for the atom, Coulomb correlations between the free electron and the atom are quite significant with resonances being found at the bound excitation energies. At longer wavelengths, the first-order correlation effects become overwhelmed by the increasing lowest-order continuum-to-continuum dipole matrix elements. It was found that certain second-order correlation effects are quite significant at both the short and long wavelengths. These second-order polarization-type diagrams represent the leading terms in the construction of the full optical potential for the free electron. We plan to extend this work in the future to investigate the free-free absorption process in the presence of molecules.

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