Unusual radiation effects from atoms in gases and plasmas*

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New interesting effects arise, when three-level atoms interact with the plasma, laser field, or a heat bath. If the atoms inside the plasma are excited by the polarized laser pulse or an electron beam, the line wings of the radiation are completely polarized, while the line core is only partially polarized. The Einstein coefficients are generalized to the case, when the atomic decay channels are subject to quantum interference. Such atoms may develop nonzero quantum coherence between upper levels, if they are in contact with the heat bath in thermal equilibrium. This coherency changes the emissivity of the medium and hence can be experimentally observed. In the case when three-level atoms interact with the laser field, spontaneous emission can be suppressed over the whole frequency spectrum. © *1999 American Institute of Physics.* [S1070-664X(99)93105-1]

I. INTRODUCTION

We present a study of different radiation effects, which are predicted to occur when atoms interact with the plasma, laser field, or a heat bath. These effects arise only, if the atoms are modeled by a three-level scheme. The presence of the third level proves to be crucial.

First, we examine the problem of polarization of the atomic radiation from the plasma. Suppose, that atoms are excited by the polarized laser pulse or an electron beam. We show, that the wings of the atomic line are completely polarized, while the core of the line is only partially polarized.[1] This effect may be responsible for a recent experimental results on the polarization of the x-ray radiation from the *Z*-pinch experiment reported by Oks.[2] In this experiment Oks *et al.* observed a current of electrons with energy 100 eV, which is capable of exciting preferentially π states. Thus, the differences in degree of polarization should be attributed both to the asymmetrical distribution of the turbulent fields and the differences in the initial populations.

Second, we consider decay of a three-level atom and show, that the Einstein coefficients should be generalized. We find, that when the quantum numbers of the upper atomic levels meet certain constraints, the relaxation operator acquires nondiagonal elements. The physical reason for these new elements is the quantum interference between different decay channels of the atom. This interference may lead to new phenomena such as lasers without inversion,[3,4] spectral line narrowing,[5,6] and line elimination.[7]

We find,[8,9] that besides the well-known term $\sqrt{A_2 A_3}$, the relaxation operator also should have a term due to Lamb shifts of the levels. The interaction of such atoms with the photon fields of different statistics is not well understood, even when the term due to Lamb shifts is not important. We study the interaction of these atoms with the laser field and the heat bath in thermodynamic equilibrium.

We show, that when such atoms are in equilibrium with the heat bath quantum interference between decay channels leads to a nonzero coherency between upper levels. This coherency can be observed in principle, because it makes a nonzero contribution to the emissivity of the medium. As a result, the emissivity has a maximum in the wing of the spectrum as well as a zero point at a frequency close to the resonance. The unusual maximum in the wing is present also in the case of a two-level atom. It was observed[10] in a recent experiment on sodium vapors by Leonov *et al.* We find,[11] however, that as a result of quantum interference the height of this maximum is twice as much as compared to the case of no interference.

Another interesting effect should occur, if the three-level atoms interact with the laser field. We predict, that the spontaneous emission can be suppressed over the whole range of frequencies at a certain amplitude of the laser field. This effect can be very useful for development of the x-ray lasers, since it would allow to effectively lock the population in the upper levels just by illuminating the medium with the laser field of certain intensity. We performed calculations with the toy model, assuming that decay channels may interfere. There was a recent experiment,[12] where a real molecular system described by this model was found. It remains to be seen, if this effect can be observed in atoms.

The paper is organized as follows: in part II we study the polarization of atomic radiation coming from the plasma; in part III we explain why the Einstein coefficients should be generalized and present the answer for the relaxation operator; in part IV we analyze the role of quantum interference in the case of thermodynamic equilibrium; in part V we calculate the intensity of the spontaneous emission and show, that it is suppressed at a certain amplitude of the laser field; in part VI we present a conclusion.

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II. POLARIZATION OF ATOMIC RADIATION

Suppose we excite an atom into one of the magnetic sublevels with the help of the electron beam or a polarized laser pulse. In the presence of the electric microfield of the plasma atomic electron will experience transitions between different sublevels. The more time that elapses between the excitation and emission of the photon, the more likely the photon will be unpolarized. Let us consider this in more detail.

Suppose a distribution of microfields is isotropic, but sharply peaked around a given amplitude. We find that emission in the line core is almost unpolarized, while the line wings are almost completely polarized. This can be understood from the uncertainty principle: Atoms radiating in a time $\Delta\tau$ emit into the line wings at a characteristic frequency detuning $\Omega \equiv \omega - \omega_0 \sim 1/\Delta\tau$, where ω_0 is the resonant frequency. Let T be the characteristic time for mixing of the magnetic sublevels in the presence of the plasma microfields, which we assume to be less than the decay time. For $\Delta\tau \ll T$, an atom does not have time to change its state before it decays; hence, it emits a photon of polarization corresponding to the preferentially excited sublevel. On the other hand, for $\Delta\tau \geqslant T$, the atomic states are mixed due to plasma microfields prior to the line emission. Thus, emission into the wings, arising from short radiation times, should be polarized, whereas emission into the core, arising from long radiation times, should be much less polarized. Of course, in a plasma, if the distribution of the microfields is not sharply peaked around a given amplitude, then the observed radiation would be an averaged effect over a distribution of amplitudes. As shown below, this average can retain important features of the sharply peaked case.

Incidentally, this effect should occur also in the presence of the polarization effects discovered in the Z-pinch experiment reported by Oks,[2] because the axial current should preferentially excite π states. Thus, the differences in degree of polarization between the core and wings could be attributed both to the turbulent fields and the differences in the initial populations.

In order to gain insight into spontaneous emission of an atom in stochastic plasma fields, consider first the case of a constant electric field, where an atom is excited into one magnetic sublevel. Suppose the atomic structure depicted in Fig. 1. Transitions occur between the degenerate upper level $|J=1,\ m=0,\pm 1\rangle$ and the lower level $|J=0,\ m_j=0\rangle$. The external electric field couples the upper level $|J=0,\ m=0\rangle$ to the $\sigma = |J=1, m=0,\pm 1\rangle$ levels. Let the z axis be along the direction of a circularly polarized laser pulse. (The case of linear polarization is handled similarly.)

Projecting the Schrodinger equation onto eigenfunctions of the angular momentum, we get a system of linear differential equations for the amplitudes

$$i\frac{da_\sigma}{dt} = V_{\sigma u} a_u, \quad i\frac{da_u}{dt} = \sum_\sigma V_{u\sigma} a_\sigma, \tag{1}$$

where $V_{u\sigma} = VE^*_\sigma = d_{u\sigma} E E^*_\sigma$ is a matrix element of the interaction with electric field, and E and E_σ are the amplitude and dimensionless spherical component of the electric field, respectively.

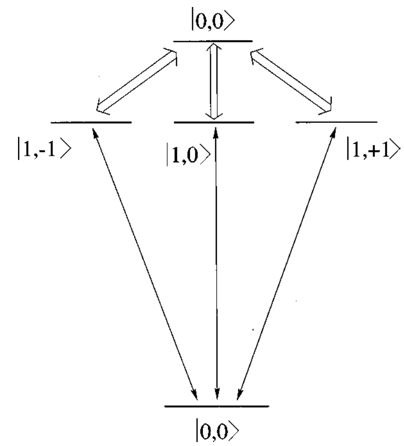

FIG. 1. Double lines represent mixing of levels due to static electric field; thin lines correspond to spontaneous decay of σ levels. Upper levels are degenerate.

For initial conditions $a_\sigma(t=0) = \delta_{\sigma\alpha}$, Eq. (1) has the solution

$$a_\sigma = E^*_\alpha E_\sigma (\cos Vt - 1) + \delta_{\sigma\alpha} \tag{2}$$

$$a_u = -iE^*_\alpha \sin Vt, \tag{3}$$

where α stands for the polarization of the pulse.

For a right-hand circularly polarized laser pulse $\alpha=1$, Eqs. (2) and (3) give the expectation value of the dipole moment of the transition between upper and lower levels as

$$\mathbf{d} = \sqrt{2} d [(E_x^2 \mathbf{e}_x + E_x E_z \mathbf{e}_z)(1-\cos Vt)\cos\omega_0 t - (\mathbf{e}_x \cos\omega_0 t + \mathbf{e}_y \sin\omega_0 t)], \tag{4}$$

where the electric field is assumed to lie in the $x-z$ plane. Note that the tip of the dipole moment vector describes an elliptical path in time $1/\omega_0$; the plane of this path, initially in the $x-y$ plane, oscillates around y axis with frequency $1/V$. The electric field behaves similarly, as shown in Fig. 2.

Consider now a laser pulse say of right-hand circular polarization, exciting an atom that is subjected to the stochastic microfields of a plasma. The line formation can be treated in two limits, impact and quasistatic,[13] corresponding to emission into the line core and line wings. Frequency

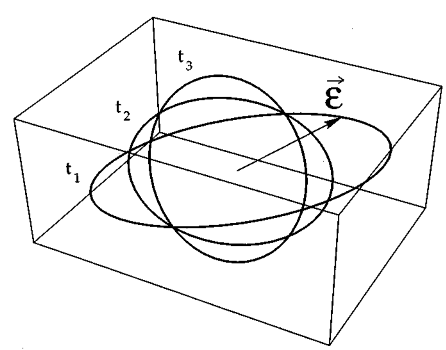

FIG. 2. Polarization of radiation when $E_z \neq 0$.

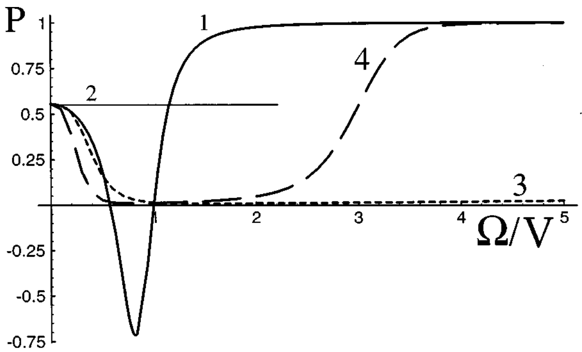

FIG. 3. Degree of circular polarization vs frequency: curves 1 (quasistatic) and 2 (impact) correspond to $P(\Omega)$ at one particular value of the microfield, V, with $\Gamma/V=0.01$; For curves 3 and 4, V represents an averaged quantity over the corresponding microfield distribution, Holtzmark or Gaussian, plotted for the case $\Gamma/V=0.01$.

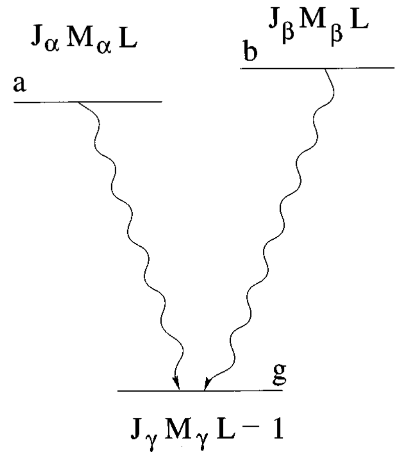

FIG. 4. Upper levels a and b have the same parity L, while the parity of the lower level g differs by 1, to allow the dipole coupling.

detuning Ω less than the Weisskopf frequency ω_W[14] corresponds to the line core, while detuning greater than ω_W corresponds to the line wings.

The atomic spontaneous emission spectrum can be written as[15]

$$I(\omega)=2h\omega Re[iG_\sigma \rho_{g\sigma}(s=-\Omega)], \tag{5}$$

where G_σ is the matrix element of the Hamiltonian of the interaction between an atom and the spontaneous field, with polarization σ and frequency ω, and where $\rho_{g\sigma'}(s=-\Omega)$ is the Laplace transformed atom+field density matrix element, $\rho_{g\sigma}(t)$, evaluated at $s=-\Omega$. In order to find spontaneous emission spectrum, one has to solve equations for the atomic as well as atom+field density matrices. The detailed calculations can be found in Ref. 1. Here we present only the final result of these calculations, Fig. 3.

The curve 1 in Fig. 3 corresponds to the calculations, assuming that the atoms are subject to the electric microfield which has a random direction but a fixed amplitude at any given point in space. So, the density matrix calculations support our qualitative arguments: the line wings are completely polarized, while the line core is only partially polarized. Note also a prominent dip in curve 1, which means that, at the frequency $\Omega\sim V$, the emitted photon is predominantly counterpolarized, namely, left-hand polarized. The fact that I_- is greater than I_+ at the frequency detuning $\Omega\sim V$ arises from Stark oscillations of $\rho_{g\sigma}$ at frequency V, with initial conditions $\rho_{\sigma\sigma'}(t=0)=\delta_{\sigma 1}\delta_{\sigma' 1}$.

In nonequilibrium plasmas where the distribution of the microfield amplitudes is sharply peaked around some given amplitude, the curve 1 in Fig. 3 should give a realistic answer. Otherwise, it should be averaged with respect to the microfield distribution. The result of such averages with respect to the Holtzmark and Gaussian distributions is presented by curves 3 and 4. For convenience, we give the formula for the Holtzmark distribution:[16]

$$W(\beta)=\frac{2\beta}{\Pi}\int_0^\infty \exp(-y^{3/2})y\sin\beta y\,dy, \tag{6}$$

where β is the normalized field.

Fluctuations in the field amplitude in equilibrium plasma described by the Holtzmark and Gaussian distributions are due to the particle and wave fields correspondingly. As one can see from Fig. 3, averaging with respect to the Gaussian distribution retains both the polarization in the wings and the pronounced minimum. The averaging with respect to the Holtzmark distribution leaves intact only partial polarization in the core, while the polarization in the wings becomes muted. This difference between the influence of particle and wave fields on the degree of polarization could serve to distinguish between the relative intensities of long wavelength and short wavelength stochastic microfields.

III. GENERALIZED EINSTEIN COEFFICIENTS

Consider a purely radiative relaxation of an atom modeled by a three-level scheme in Fig. 4. We show, that because of the presence of the third level the relaxation matrix may acquire nondiagonal elements, i.e., to describe the radiative relaxation of the three-level atoms it is not sufficient to specify only the usual Einstein coefficients.

This can be shown qualitatively as follows. To find the rate of spontaneous emission we have to consider the interaction of the atomic levels with the infinite number of modes of the vacuum electromagnetic field. Each mode is characterized by the wave vector and by the polarization vector. Let us pick one such mode. Provided the restrictions on the quantum numbers of the atomic levels are met,[8,9] this mode can interact with both upper levels at the same time. When summed over all possible quantum modes, this interaction will clearly lead to the coupling between the upper levels, which can be described by the nondiagonal elements of the relaxation operator.

One can also describe this effect in different terms. In a three-level atom there are two channels of decay, $a\rightarrow g$ and $b\rightarrow g$. Since we are usually interested only in the outcome of the decay process, we make measurements of the spectral distribution of the photons emitted in the process, rather than electron populations directly. Hence, two decay channels are free to interfere resulting in the nondiagonal elements of the relaxation operator.

The result of the full calculations[8,9] is given by the following expression:

$$\Gamma_{aa}=(A_a+i\Delta_a),\quad \Gamma_{bb}=(A_b+i\Delta_b),\tag{7}$$

$$\Gamma_{ab}=(\sqrt{A_aA_b}+i\sqrt{|\Delta_a\Delta_b|}).\tag{8}$$

Here A_a and A_b are the usual Einstein coefficients, while the Δ_a and Δ_b are the Lamb shifts. We see, that nondiagonal element indeed appear. It is equal to the mean geometric average of the Einstein coefficients plus the nonlinear term from Lamb shifts.

It is interesting to consider how the atoms with such relaxation operators behave in different environments. First, consider the case, when they are in thermodynamic equilibrium with the heat bath.

IV. QUANTUM INTERFERENCE AND THERMODYNAMIC EQUILIBRIUM

The case of thermodynamic equilibrium is not as simple as it may seem, even for the case of a two-level atom. The usual calculations based on the atomic model with infinitely sharp levels lead to an incorrect result, with the atomic frequency of transition ω_0 in the exponent, $\exp(h\omega/T)$, rather then the current frequency of the photon ω as demanded by the Planck formula.

To obtain the correct black body spectrum, one should consider levels with a nonzero width, which implies infinitely many atomic virtual levels. Therefore, one has to consider an interaction between infinitely many atomic oscillators and infinitely many field oscillators, something that can be successfully accomplished with the help of the field theory methods. We use the Keldysh–Korenmann approach[17,18] and solve the system of equations for the atomic and photon Green functions.[11] Kinteic atomic Green functions in case of thermodynamic equilibrium between three-level atoms and the photon gas are given by the following formulas:

$$G^{-+}_{uu'}(\omega)=-\frac{1}{e^{\omega/T}+1}2\pi i a_{uu'}(\omega),\tag{9}$$

$$a_{uu'}(\omega)=\frac{\sqrt{A_uA_{u'}}\Omega_u\Omega_{u'}}{(\Omega_2\Omega_3)^2+\frac{1}{4}(\Omega_2A_3+\Omega_3A_2)^2}.\tag{10}$$

Here u and u' stand for upper atomic levels 2 and 3. The physical meaning of these functions is close to the meaning of the distribution function so often used in plasma physics, except that these functions incorporate quantum effects. In fact, the diagonal elements, $G_{uu}(\omega)$, give the probability for the atomic electron to occupy the virtual level of frequency ω, which is characterized by the quantum number u. The nondiagonal elements give the coherency between levels u and u'.

As we can see from Eqs. (9) and (10) three-level atoms with decay channels subject to quantum interference acquire nondiagonal elements in the atomic Green functions even in thermodynamic equilibrium. The spectrum of the radiation, however, is given by the Planck formula, as it should in the case of equilibrium.

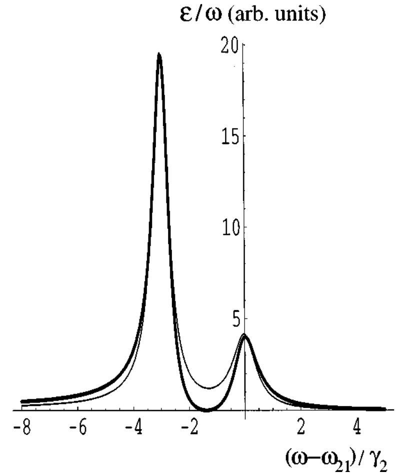

FIG. 5. Zero point in the emission spectrum. Frequency is normalized to the width γ_2 of the level 2. Atomic parameters are: $\omega_{32}/\gamma_2=-3$, $\omega_{21}/\gamma_2=100$, $T/\gamma_2=5$, $d_{31}/d_{21}=\sqrt{2}$, $\omega_{31}/\omega_{21}=0.7$. Thick and thin lines correspond to spectra with and without interference.

Yet, these nondiagonal elements give nonzero contribution to the spontaneous source of the radiation. The physical meaning of this source can be most easily understood from the simplified equation of the radiation transport:

$$(\Omega\cdot\nabla)J(\omega,\mathbf{r})=-k(\omega)J(\omega,\mathbf{r})+\epsilon(\omega),\tag{11}$$

$$J(\omega,\mathbf{r})=\frac{\epsilon(\omega)}{k(\omega)}.\tag{12}$$

The left-hand side of Eq. (11) describes the free propagation of photons in the steady state, while the right-hand side consists of sources and sinks. The source term, $\epsilon(\omega)$, is the spectral density of the spontaneous emission, while the sink term, $k(\omega)J(\omega,\mathbf{r})$, is the product of the spectral density of the radiation $J(\omega,\mathbf{r})$ and the absorption coefficient $k(\omega)$ corrected for the induced emission. In equilibrium, if the optical depth of the heat bath at a given frequency ω is $\tau(\omega)\gg 1$, then the photon of such frequency was reemitted many times, and therefore the intensity of the radiation is given by the usual Kirhoff law, Eq. (12), which is the black body spectrum. If, however, $\tau(\omega)\leqslant 1$, then the distribution of the radiation is given by the spontaneous source $\epsilon(\omega)$.

We found the contribution of the nondiagonal elements of the atomic Green functions $G^{-+}_{uu'}(\omega)$ to this spontaneous source.[11] It is given by

$$\epsilon(\omega)=\frac{2}{3}\frac{\omega^4}{4\pi c^3}N_F(\omega)\sum_{uu'}d_{u1}d_{u'1}a_{uu'}(\omega),\tag{13}$$

where d_{u1} and $d_{u'1}$ are the dipole moments between the upper and ground levels and $N_F(\omega)$ stands for the Fermi distribution. The plot of this function is given in the Figs. 5 and 6.

We see, that the spectrum acquires a zero point at a certain frequency, Fig. 5. Also, quantum interference leads to a twofold enhancement in the red wing of the spectrum.

In the case of two-level atoms, when there is no quantum interference between decay channels a similar red wing was recently observed in the experiment by Leonov *et al.*[10]

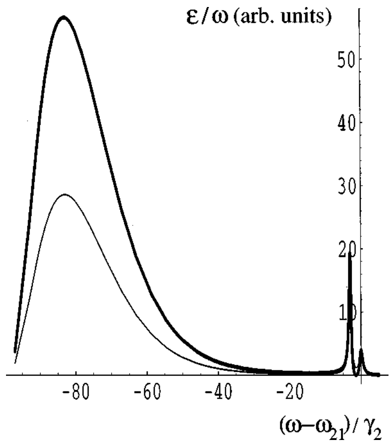

FIG. 6. Enhancement of the red wing due to quantum interference. All conventions are the same as in Fig. 5.

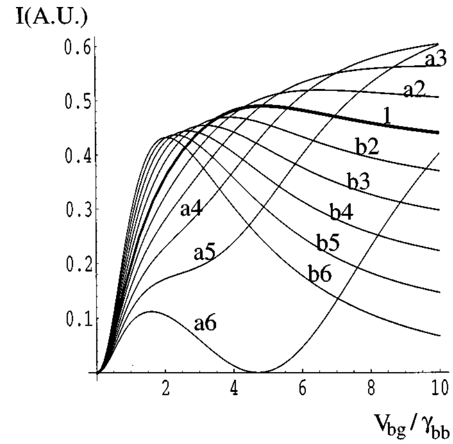

FIG. 7. Intensity of spontaneous emission vs V_{bg}/γ_{bb} for laser detuning $\omega_{Lb}/\gamma_{bb}=-8$. Curves 1, $a2..an..a6$ correspond to successive values of $q=0.2(n-1)$, curves $b2..b6$ are plotted for $q=-0.2(n-1)$ with $n=2,..,6$. Atomic parameters normalized to γ_{bb} are: $\omega_{ab}=10$, $d_{bg}/d_{ag}=\sqrt{2}$, $\omega_{bg}/\omega_{ag}=0.7$.

V. ATOM IN THE LASER FIELD

Now let us consider the interaction of the three-level atoms with the laser field. It is governed by the system of equations for the atomic density matrix

$$i\dot{\rho}=[H,\rho]+i\Gamma[\rho], \quad H=H_0+H_r+H_i. \tag{14}$$

Here H_0 is the atomic Hamiltonian and H_r is the free radiation field Hamiltonian. The interaction Hamiltonian H_i describes the interaction of the atom with the laser field, and the relaxation operator $\Gamma[\rho]$ is given[8] by Eqs. (7) and (8). The total rate of spontaneous emission from the upper levels in the steady state is given by the following expression:

$$I\propto\rho_{aa}\Gamma_{aa}+\rho_{bb}\Gamma_{bb}+2qRe[Re[\Gamma_{ab}]\rho_{ab}], \tag{15}$$

The first two terms in this equation give the usual decay of the populations due to the radiative transitions. The third term arises because of the quantum interference between different decay channels, which is described by the nondiagonal elements of the relaxation operator.

We showed, that for two decay channels to interfere in free space the quantum numbers of the upper levels have to meet certain constraints. In case, when they do not meet this constraint and there is no interference in free space, we find,[9] that the interference still can take place, if the atom is inside the cavity or the dielectric medium. The q coefficient in front of the third term mimics the influence of the cavity or the dielectric medium.

We solved[9] Eq. (14) in the steady state regime and found the analytical expression for the intensity of spontaneous emission, $I(V)$. The plot of this function is presented in the Fig. 7.

The most striking feature of the function $I(V)$ is that it is equal to zero at a certain amplitude of the laser field as can be seen from the Fig. 7. This zero occurs, when the q coefficient is equal to 1, which corresponds to the case when there is no resonator and the atom interacts only with the laser field. This result means, that the spontaneous emission is suppressed over the whole frequency spectrum and not just at a certain frequency. We found[8,9] an analytical expression for the laser field amplitude, when this suppression occurs:

$$V^2=\left(\frac{\omega_b}{\omega_a}\right)^{3/2}\frac{\omega_{ba}\left(\omega_{Lb}+\omega_{La}\left(\frac{d_{bg}}{d_{ag}}\right)^2\left(\frac{\omega_b}{\omega_a}\right)^{3/2}\right)}{\left(1-\left(\frac{\omega_b}{\omega_a}\right)^{3/2}\right)\left(1+\left(\frac{d_{bg}}{d_{ag}}\right)^2\left(\frac{\omega_b}{\omega_a}\right)^{3/2}\right)}. \tag{16}$$

VI. CONCLUSION

In this paper we showed, that new interesting effects arise when the atom modeled by the three-level scheme interacts with different environments. If the atoms inside the plasma are excited by the polarized laser pulse or an electron beam the wings of the atomic line should be polarized, while the line core should be only partially polarized. This polarization dependence is very sensitive to the distribution of the electric microfields of the plasma and may serve as a useful diagnostic tool to distinguish between the influence of particle and wave fields.

We generalized the Einstein coefficients to the case, when the three-level atom has decay channels subject to quantum interference. We considered the interaction of such atoms with the heat bath in thermal equilibrium and found, that the quantum coherency may develop between upper levels. This nonzero coherency changes the emissivity of the medium, which in principle can be observed.

We showed, that when three-level atoms interact with the laser field, spontaneous emission can be suppressed over the whole range of frequencies. This effect can be used to obtain a population inversion on the atomic transition which has a large value of the Einstein coefficient.

ACNOWLEDGMENTS

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