EFFECT OF ARGON BUFFER GAS AND THE DIMER COMPONENT ON THE OPTICAL PROPERTIES OF SODIUM VAPOR

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We have theoretically studied the nature of strong optical losses in the red region of the spectrum that occur in sodium vapor. We take into account the effect of the Ar buffer gas and Na\(_2\) dimers on the resulting absorption of light by the system. Based on the proposed approach, we determine the interatomic distance in the dimers and achieve good agreement with quantum calculations and direct measurements. We show that intrinsic transitions in atomic argon may be the reason for the appearance of a broad absorption peak in the range 700–1000 nm.

Keywords: alkali metal vapor, "unexpected" losses, dimer, absorption spectrum, dipole resonance.

Introduction. An unresolved problem today is the origin and structure of some regions of the absorption and transmission spectra of alkali metal vapor. Thus an important problem is the nature of intense emission in the near IR region of the spectrum of high-pressure sodium gas-discharge lamps. Unproductive losses going toward IR emission, which may be as high as 20% to 30% of the total energy balance in sodium lamps, have been called "unexpected" losses for a long time and to date have not been even qualitatively explained within any theoretical model.

Intensity IR emission has been detected in the thermoluminescence spectra of dense potassium [1] and sodium [2] vapors, where argon acted as the buffer gas. The question of the origin of the intense IR emission of dense alkali metal vapors remains unanswered at this time. The theoretical model in [2], constructed assuming the quasistatic approximation,\(^{**}\) describes the experimental data well close to resonance but does not describe the observed intense luminescence in the IR region. Its possible appearance is attributed in [2] to the effect of metal clusters, which can be formed under nonequilibrium conditions both in heated cuvets of the "heat pipe" type [3] and in high-pressure sodium lamps. This question is also quite interesting from the standpoint of designing cluster lamps [4], since optimal control of the spectral properties of the lamps seems possible. In this case, in [5, 6] the effect of a microdroplet component on the optical transmission spectrum of sodium vapor was analyzed, but the results obtained do not support the hypothesis that clusters play an important role in forming the absorption lines in the red and near IR regions. One mechanism for forming the absorption spectrum of alkali metals is also the interaction between radiation and atomic dimers, where as shown in [6], the concentration of the latter in dense vapors is quite significant.

We should point out that available papers neglect the effect of intrinsic absorption peaks of the buffer gas on the resulting optical properties of the system. The authors of [7] noted the disappearance of absorption in the red region when the buffer gas was removed (argon in this case), in the spectrum of which the strongest transitions are located specifically in the range 700–1000 nm. The experimental data presented in [6] suggest that the pressure and type of buffer gas have a quite strong effect on the optical properties of the studied medium: absorption increases as the buffer gas pressure increases, and proves to be significantly greater for argon than for helium. Accordingly, this work was devoted to studying the effect of the buffer filler and also the dimer components arising in dense metal vapors on the resulting absorption spectrum of the system as a whole. Based on the proposed approach, we obtained simple expressions for the effective refractive index for sodium vapor, and also determined the parameters of the Na\(_2\) mole-

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\(^{**}\)In calculation of the line shape, the particles are considered stationary, i.e., Doppler broadening of the line is neglected.
cules. We obtained good quantitative agreement with the experimental data in [5], and also with the results of a quantum mechanical study of sodium dimers [8]. It was shown that peaks in the blue region, like the strong absorption in the near IR region, are due to Na₂ molecules while the "unexpected" losses in the red region can be explained by the presence of argon.

**The Model. Absorption in Sodium Vapor.** Let us consider some arbitrary medium, in the general case consisting of several types of atoms, occupying a volume V, on which is incident an external electromagnetic wave with intensity \( E_0(\mathbf{r}, t) \). Let us make use of the coupled dipole method, which we have repeatedly used previously to study nanosized objects and structures and which has shown good agreement with both experimental data and *ab initio* exact numerical calculations [9–11]. Within this approach, the field at an arbitrary point in the medium is written in the form [12, 13]:

\[
E(\mathbf{r}, t) = E_0(\mathbf{r}, t) + \sum_j \nabla \times \nabla \times \frac{\mathbf{p}_j (t - |\mathbf{r} - \mathbf{r}_j|/c)}{|\mathbf{r} - \mathbf{r}_j|}, \tag{1}
\]

where \( \mathbf{r} \) is the radius vector of the observation point; the \( \mathbf{r}_j \) are the radius vectors of the atoms; \( c \) is the speed of light in a vacuum; \( \mathbf{p}_j \) is the induced dipole moment of the atom labeled \( j \); derivatives are taken with respect to the coordinates of the observation point. Let us assume that the dipole moments of the atoms in the system are linearly dependent on the field:

\[
\mathbf{p}_j(\mathbf{r}, t) = \alpha_j E_0(\mathbf{r}, t). \tag{2}
\]

In the case when the average distance between atoms is much smaller than the wavelength, the summation in (1) can be replaced by integration [12]. Then for a multicomponent medium, we obtain:

\[
E(\mathbf{r}, t) = E_0(\mathbf{r}, t) + \sum_j \int_V \nabla \times \nabla \times \frac{N_j \alpha_j E (t - |\mathbf{r} - \mathbf{r}'|/c)}{|\mathbf{r} - \mathbf{r}'|} dV. \tag{3}
\]

Here \( N_j \) and \( \alpha_j \) are the concentration and polarizability of the \( j \)-th type of atoms. Using the standard procedure for taking the operator \( \nabla \times \nabla \times \) out from under the integral sign, we can obtain the extinction theorem [12] and the equivalent of the Lorentz–Lorenz formula for the refractive index of a multicomponent system \( n_{\text{eff}} \) [14]:

\[
\frac{n_{\text{eff}}^2 - 1}{n_{\text{eff}}^2 + 2} = \frac{4\pi}{3} \sum_j N_j \alpha_j. \tag{4}
\]

The major components of the studied system are atomic argon and sodium, and also some number of sodium dimers for which we need a description of the optical properties in the following. In this paper, we consider the interaction between the sodium dimers and the electromagnetic field within the semiclassical approach proposed and tested earlier in [13–15]. Let us write the field created by the dimer at any point in space according to (1) in the form:

\[
E(\mathbf{r}, t) = E_0(\mathbf{r}, t) + \sum_{j=1}^2 \nabla \times \nabla \times \frac{\mathbf{p}_j (t - |\mathbf{r} - \mathbf{r}_j|/c)}{|\mathbf{r} - \mathbf{r}_j|}, \tag{5}
\]

where the summation is carried out over the atoms making up the dimer. The dipole moments of the atoms in the system, \( \mathbf{p}_j \), are defined by expression (2), while their polarizabilities are defined in the classical form [12]:

\[
\alpha_j = \frac{e^2}{m_e} \sum_{i=1}^2 \frac{f^{(i)}_{\text{Na}}}{\omega_i^2 - \omega^2 - i\gamma^{(i)}_{\text{Na}}}. \tag{6}
\]

Here \( i \) is the numerical label of the line in the spectrum of the atom; \( m_e \) and \( e \) are the electronic mass and charge; \( f^{(i)}_{\text{Na}}, \omega_i, \gamma^{(i)}_{\text{Na}} \) are respectively the oscillator strength, the resonant frequency, and the decay constant of the \( i \)-th transition. The decay constant for the atoms in the dimer (and consequently their polarizability), however, may be different from the value for a solitary atom, therefore