Temperature dependence of YbHe continuum emission spectra

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We have observed laser-excited continuum emission spectra in the far wings of the $6s^2\,{}^1S_0$–$6s6p\,{}^1P_1$ resonance line of Yb broadened by He at seven different temperatures (405–560 °C). We find that the YbHe molecular excited state $6s6p\,{}^1\Sigma$ is repulsive.

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I. INTRODUCTION

Since pioneering work by Hedges *et al.* [1], several groups have investigated far-wing spectra of the resonance lines of alkali-metal and alkaline-earth-metal atoms broadened by rare gases (RG's) [2–14]. Their main concerns were to obtain information about interatomic potential-energy curves as well as about collision dynamics for the metal-RG systems.

In the course of our systematic studies on optical transitions of quasimolecules formed during thermal collisions in metal-vapor–rare-gas mixtures [15–18], we have reported continuum absorption spectra observed in far wings of the $6s^2\,{}^1S_0$–$6s6p\,{}^1P_1$ resonance line of Yb broadened by He, Ne, Ar, Kr, Xe, and Yb [19]. We obtained approximate difference-potential energies between the $6s^2\,{}^1\Sigma$ and $6s6p\,{}^1\Sigma$ Yb-RG molecular states from our absorption spectra with the aid of unified Frank-Condon (UFC) line-shape analysis of Szudy and Baylis [20]. The information about the potential energies for the respective upper and lower molecular states, however, could not be obtained from our absorption spectra.

In the present work, we measure the temperature dependence of continuum radiation in far wings of the resonance line of Yb broadened by He, hoping to obtain information about the potential energies of the respective upper and lower molecular states.

II. EXPERIMENT

A schematic diagram of our step is illustrated in Fig. 1. The Yb vapor was contained in a heat-pipe fluorescence cell with an inner diameter of 20 mm. Its central portion, 80 mm in length, was heated to 405–560 °C. 100–700-Torr He gas was added as a perturber. The Yb vapor in the cell was irradiated with the laser whose wavelength was tuned on the $6s^2\,{}^1S_0$–$6s6p\,{}^1P_1$ resonance line of Yb at $\lambda=399$ nm. A frequency-doubled 10-Hz Q-switched Nd:YAG laser (where YAG represents yttrium aluminum garnet) was used to pump a pulsed dye laser operated with DCM laser dye in an oscillator-amplifier arrangement. The dye-laser output at $\lambda=638$ nm was then mixed with the $\lambda=1.06$ μm Nd:YAG fundamental in an angle-tuned KD*P crystal to produce the $\lambda=399$ nm radiation. The unwanted wavelengths were separated with a wavelength separator having two Pellin-Broca prisms. The laser pulse energy was ~ 100 μJ with a pulse length of ~ 5 ns. The laser was softly focused in the heating zone of the fluorescence cell. To observe a backward fluorescence, we used a pierced mirror. The fluorescence reflected by the pierced mirror was collimated and focused with two quartz lenses (50 mm in diameter and 300 mm in focal length) onto the entrance slit of a 500-mm Czerny-Turner spectrometer equipped with an 1800-grooves/mm grating. The entrance slit width was 50 μm. The dispersed fluorescence was detected with a spectroscopic multichannel analyzer which consists of a photodiode-array detector and a gated image intensifier (Princeton Instruments D/SDIA-700). This detector covered at one time the wavelength range $\lambda=390$–406 nm in which the resonance line of Yb at $\lambda=399$ nm was included. The gate was set to open ~ 30 ns after the excitation laser in order to eliminate the scattered laser light. The gate duration was ~ 200 ns. The spectral resolution was ~ 0.1 nm.

During the measurement, the temperature of the fluorescence cell was monitored on the outer wall of the cell using a Chromel-Alumel thermocouple and was regulated within 1 °C with a programmable temperature controller. In a separate experiment, we measured the temperature difference between the outer wall of the cell and the area where the fluorescence was emitted by inserting another Chromel-Alumel thermocouple into the cell, and found that the difference was 15–20 °C depending on the temperature.

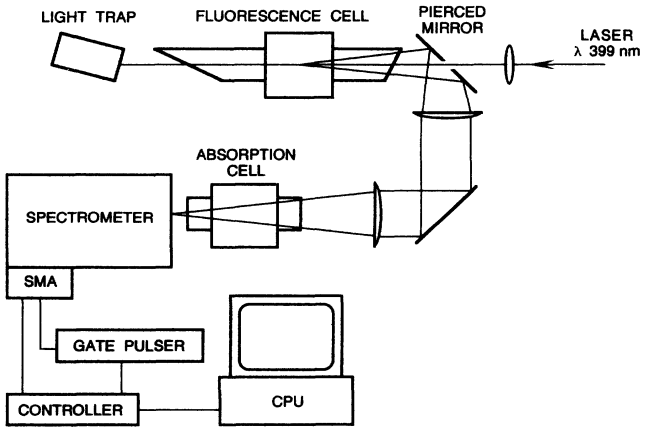

FIG. 1. Schematic diagram of the experimental setup.

A measurement of the weak far-wing emission spectra required suppression of leakage light of the line core. For this purpose, another heat-pipe cell with an inner diameter of 20 mm and a heating length of 80 mm, in which the Yb vapor was contained, was placed as an absorption filter in front of the entrance slit of the spectrometer. 100-Torr He gas was added as a buffer gas in the absorption cell. The temperature of the absorption cell was kept at ~585 °C.

III. RESULTS AND DISCUSSION

A portion of far-wing emission spectra of the resonance line of Yb broadened by He is given in Fig. 2. The temperature of the fluorescence cell was 534 °C and the He pressure was 700 Torr. We took the emission spectra similar to Fig. 2 at seven different temperatures between 405 and 560 °C and at some different He pressures. Normalizing each spectrum at $\lambda = 397.8$ nm, we found an increase in the relative intensity in the blue wing with an increase in temperature. A change of the He pressure did not change the relative intensity in both wings.

We focus our attention on the temperature dependence of the emission intensity in the blue wing. According to a classical line-shape theory [1], the emission intensity in the quasistatic wing depends on the temperature through the Boltzmann factor for the upper state of the transition:

$$I(\Delta\nu) \propto \exp[-V_u(r(\Delta\nu))/kT] , \quad (1)$$

where $r(\Delta\nu)$ is the internuclear distance responsible for radiation at detuning $\Delta\nu$ from the line center and $V_u(r)$ is the upper-state potential energy at $r(\Delta\nu)$. The upper state for the present blue-wing emission is considered to be $6s6p\ ^1\Sigma$ [19]. The emission intensities at some wavelengths are plotted as a function of $1000/T$ (K^{-1}) on a semilogarithmic scale in Fig. 3. It is evident from Eq. (1) that the slope of the fitted straight line represents

$$\Delta V_u(\Delta\nu) \equiv V_u(r(\Delta\nu)) - V_u(r(\Delta\nu_0)) , \quad (2)$$

where $r(\Delta\nu_0)$ is the internuclear distance responsible for radiation at $\Delta\nu_0$ at which the emission intensity is normalized. The resultant $\Delta V_u(\Delta\nu)$ are plotted as a function of $\Delta\nu$ in Fig. 4. The scatter among the data points

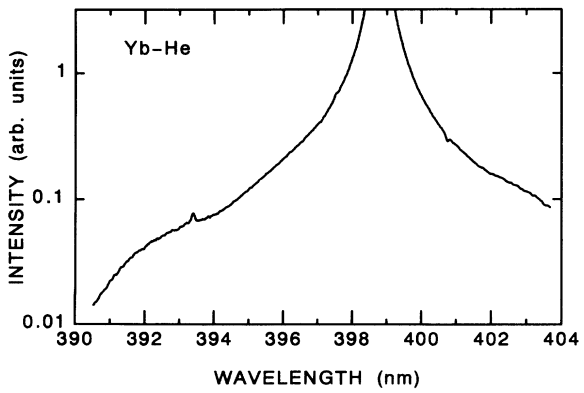

FIG. 2. A portion of the continuum emission spectrum in far wings of the $6s^2\ ^1S_0$–$6s6p\ ^1P_1$ resonance line of Yb broadened by He. The spectrum was obtained at 534 °C.

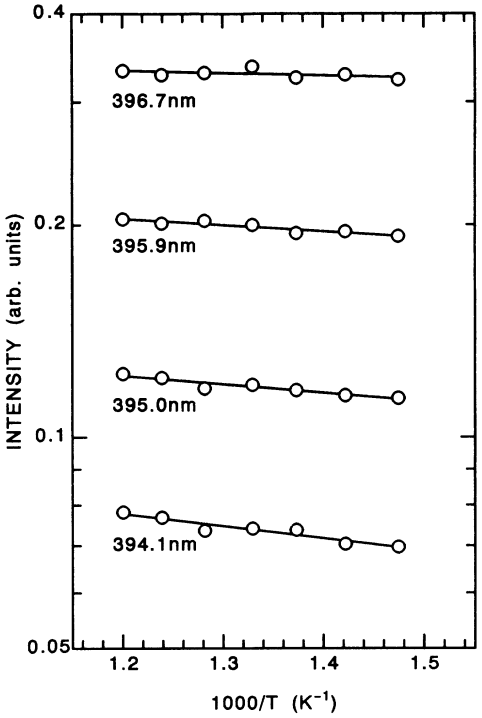

FIG. 3. Temperature dependence of the emission intensity. The relative emission intensities at some wavelengths are plotted as a function of $1000/T\,K^{-1}$ on a semilogarithmic scale.

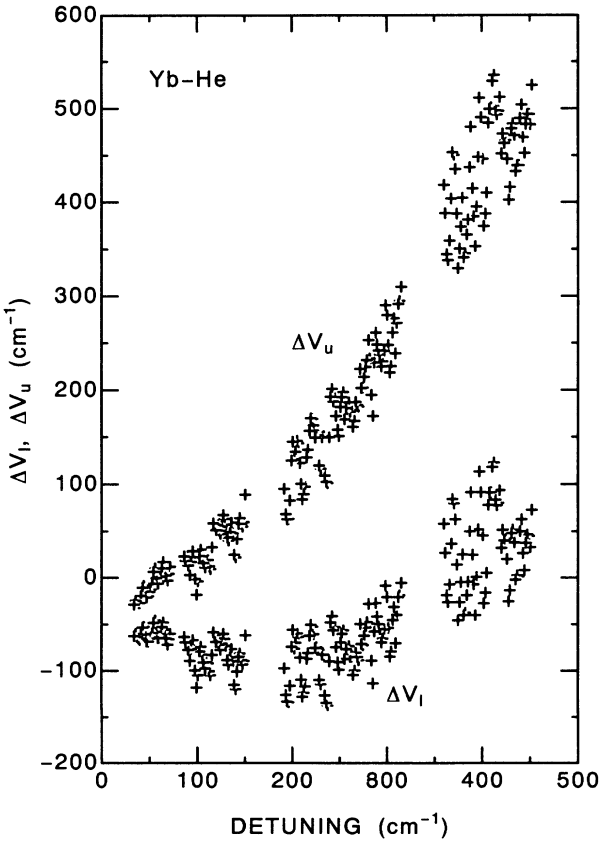

FIG. 4. $\Delta V_u(\Delta\nu)$ defined by Eq. (2) and $\Delta V_l(\Delta\nu)$ defined by Eq. (3) plotted as a function of detuning $\Delta\nu$.

reflects the statistical uncertainty. Some emission lines arising from the transitions from the higher excited states populated by the laser multiphoton excitation prevented us from measuring the temperature dependence of the continuum radiation at detuning of ~ 170 and ~ 340 cm^{-1}. The $\Delta V_u(\Delta\nu)$ increase with an increase in $\Delta\nu$ over the entire observed range of detuning, suggesting the repulsive feature of the upper state $6s6p\ ^1\Sigma$ in the region $r < r_0$. The values

$$\Delta V_l(\Delta\nu) \equiv \Delta V_u(\Delta\nu) - \Delta\nu = V_l(r(\Delta\nu)) - [V_u(r(\Delta\nu_0)) - V_u(\infty)] \quad (3)$$

are also plotted in Fig. 4. The figure clearly shows that the potential energy $V_l[r(\Delta\nu)]$ of the lower state of $6s^2\ ^1\Sigma$ is repulsive in the region for $\Delta\nu > 250$ cm^{-1}. It may be worth noting that both $\Delta V_u(\Delta\nu)$ and $\Delta V_l(\Delta\nu)$ extrapolate to ~ -50 cm^{-1}. Thus, the molecular potential energies relative to the atomic energies are the ΔV values in Fig. 4 plus 50 cm^{-1}. $V_l[r(\Delta\nu)]$ may have a van der Waals minimum of ~ 50 cm^{-1} in depth at $\Delta\nu \sim 230$ cm^{-1}, although it is not definite because of the large experimental uncertainty (say, 100 cm^{-1} including possible systematic error).

In conclusion, we have observed far-wing emission spectra of the YbHe quasimolecule. From the temperature dependence of the blue-wing emission intensity, we derived $\Delta V_u(\Delta\nu)$ and $\Delta V_l(\Delta\nu)$ in Fig. 4, illustrating the repulsive feature of the potential energies $V_u(r)$ and $V_l(r)$ for the $6s6p\ ^1\Sigma$ and $6s^2\ ^1\Sigma$ states of YbHe. We cannot convert $\Delta V_u(\Delta\nu)$ and $\Delta V_l(\Delta\nu)$ into $V_u(r)$ and $V_l(r)$ because of the lack of information on the integrated emission intensity, etc. [1].

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