Coherent hyper-Raman scattering by autoionization states of atoms excited in the plasma of an electric discharge

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A four-wave nonlinear-optics process has been detected under conditions corresponding to a hyper-Raman resonance with a transition between an autoionization state and a bound state of a copper atom. The Fano parameters are determined from the shape of the spectrum.

1. A four-wave nonlinear-optics process of a particular type has been detected experimentally for the first time in the plasma of an electric discharge. This process starts from an excited state of a copper atom and has a hyper-Raman resonance with a transition between an autoionization state and a bound state. A characteristic asymmetry of the signal has been observed. This asymmetry stems from an interference of states of the discrete spectrum with states of the continuum. A simple theory is offered for the process. This theory makes it possible to reproduce, by means of a fitting procedure, the experimental shape of the signal representing the four-wave scattering by excited states of the copper atom, in a process involving an autoionization state. The Fano parameters can thus be determined.

2. Resonances in the spectrum of a four-wave scattering involving autoionization states were first detected by Armstrong and Wynne[1] in a vapor of metal atoms. A large number of studies have been carried out on the effects of laser-induced, autoionization-like[2] resonances on the efficiency of multiphoton processes in atoms (e.g., Refs. 3 and 4).

The present experiments were carried out on excited states of a copper atom in the electric-discharge plasma of the active medium of a copper-vapor laser. We studied the spectrum of the coherent anti-Stokes scattering (CARS) $\omega_a = 2\omega_1 - \omega_2$, where ω_1 is the fixed frequency of the second harmonic of the beam from a Nd:YAG laser, and ω_2 is the output frequency of a tunable dye laser. In this case there was a resonance between the frequency ω_a (the corresponding wavelength was $\lambda_a = 4674$ Å) and the frequency of the transition between the $^4D_{5/2}$ autoionization state and the $^4F^0_{5/2}$ discrete excited state of the copper atom (see the inset in Fig. 1). A three-wave resonance of this type is associated with a hyper-Raman scattering by these states. The corresponding four-wave process is called active hyper-Raman-scattering spectroscopy (AHRS).[5,6]

3. Figure 2 shows the experimental layout. The excited copper atoms are formed in the discharge tube of a copper-vapor laser. A nanosecond CARS spectrometer is used for probing. In the course of the electric discharge, excited states of the copper atom, including autoionization states, are populated in the discharge tube.

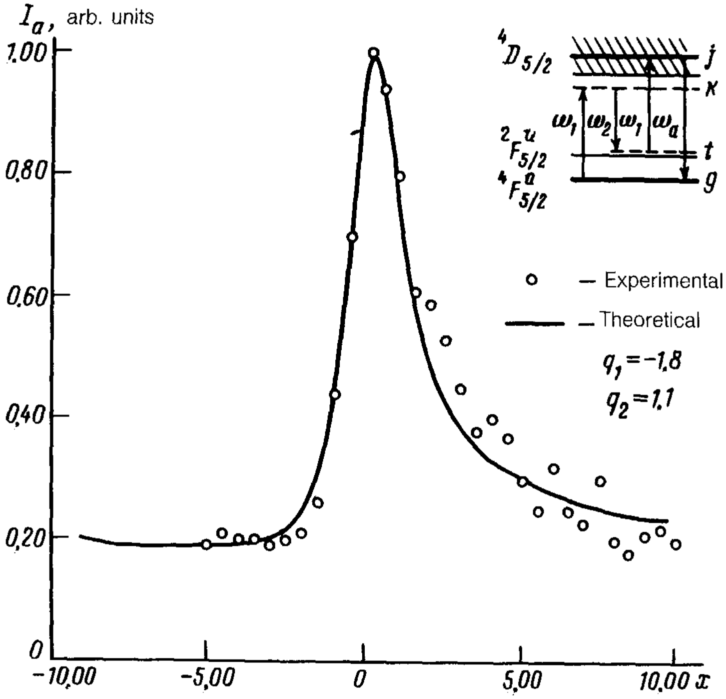

FIG. 1. Spectrum of the signal representing the active hyper-Raman scattering by copper atoms excited in the plasma of an electric discharge. The inset shows the scheme of the nonlinear-optics process under study in the case of a hyper-Raman resonance with a transition between an autoionization state and a bound state.

We studied the shape of the spectrum and the behavior of the intensity of the AHRS signal as a function of the time delay between the excitation and the probing under condition of a resonance involving an autoionization state. Figure 1 (the data points) shows the typical shape of the autoionization resonance in the AHRS spectrum.

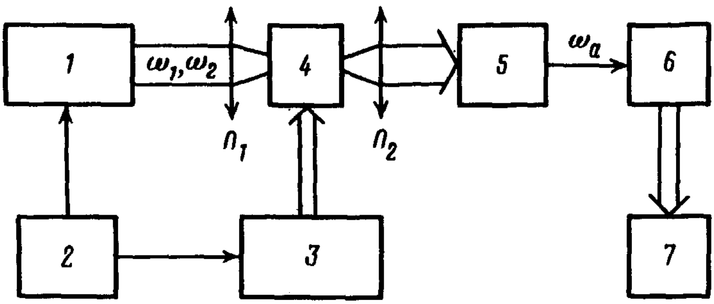

FIG. 2. Experimental layout. 1—Nanosecond CARS spectrometer; 2—unit for synchronizing the excitation and probing; 3—source for exciting the electric-discharge plasma; 4—discharge tube of the copper-vapor laser; 5—DFS-24 monochromator; 6—multichannel spectrum analyzer; 7—personal computer.

4. To calculate the shape of the spectrum of four-wave scattering by excited states of copper atoms, we work from the known expression for a cubic nonlinear susceptibility:[7]

$$\chi^{(3)} \sim \sum_{k,t,j} \frac{d_{gj}d_{jt}d_{tk}d_{kg}}{(\omega_{kg}-\omega_1)(\omega_{tg}-\omega_1+\omega_2)(\omega_{jg}-\omega_a)}. \tag{1}$$

Here d_{gj}, d_{jt}, d_{tk}, and d_{kg} are dipole moment matrix elements; g refers to the initial state, k and t to virtual states, and j to the autoionization state.

In our case there is a quasiresonance at the Raman frequency $\omega_1 - \omega_2$ with the frequency ω_{gt} of the ${}^4F^0_{5/2} - {}^2F^0_{5/2}$ transition (the detuning is 437 cm^{-1}; Fig. 1). If we retain in the sum over t only one term, with the quasiresonance denominator $\omega_{tg} - \omega_1 + \omega_2 + \Delta\omega$, we find

$$\chi^{(3)} \sim \frac{1}{\Delta\omega} \sum_k \frac{d_{tk}d_{kg}}{(\omega_{kg}-\omega_1)} \int \frac{d_g(E)d_t(E)}{E-\hbar\omega_a} dE. \tag{2}$$

By analogy with Ref. 8, we find an expression for $\chi^{(3)}$:

$$\chi^{(3)} \sim \frac{\pi N}{2\omega} d_{g\psi_E} d_{\psi_E t} \frac{q_1q_2 - i(q_2+q_1-x)}{x-i}. \tag{3}$$

Here $N = \Sigma_k [d_{tk}d_{kg}/(\omega_{kg}-\omega_1)]$ (since the four-wave scattering of interest here starts from an excited state of the copper atom, an integration over states of the continuum is assumed along with a summation over bound states in the expression for N); $x = E_s - h\omega_a/\pi|V_E|^2$;

$$q_1 = \frac{<\Phi|d|\varphi_g>}{\pi V_E^* <\psi_E|d|\varphi_g>} \quad \text{and} \quad q_2 = \frac{<\Phi|d|\varphi_t>}{\pi V_E^* <\psi_E|d|\varphi_g>}$$

are the Fano parameters;[9] φ_t and φ_g are the wave functions of bound states (in our case, φ_t is the function of the state ${}^2F^0_{5/2}$); ψ_E is the wave function of a state of the continuum; Φ is the wave function of the modified discrete state with energy E_s; and V_E is an off-diagonal matrix element.[9,10]

A comparison of the experimental data with results calculated on the shape of the four-wave scattering spectrum in accordance with expression (3) (the intensity of the anti-Stokes signal is $I_a \sim |\chi^{(3)}|^2$) makes it possible to use a fitting procedure to determine the values of the Fano parameters q_1 and q_2. A satisfactory agreement is reached with the values $q_1 = -1.8$ and $q_2 = 1.1$ (the solid line in Fig. 1). The value found for q_1 agrees within the measurement error with the value found from absorption data.[11]

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