Observation of a long-lived autoionization state in the spectrum of the gadolinium atom

G. I. Bekov, V. S. Letokhov, O. I. Matveev, and V. I. Mishin

Spectroscopy Institute, USSR Academy of Sciences
(Submitted 2 August 1978)
Pis'ma Zh. Eksp. Teor. Fiz. **28**, No. 5, 308–311 (5 September 1978)

An autoionization state with a lifetime 0.5 nsec was observed for the first time ever in selective three-step ionization of gadolinium atoms by radiation from pulsed dye lasers. A value $I = 49\ 598 \pm 5\ \text{cm}^{-1}$ was obtained for the ionization energy of the gadolinium atom.

PACS numbers: 32.80.Dz

1. The development of methods of stepwise excitation of high-lying states of atoms by radiation of pulsed tunable dye lasers has made it possible to investigate autoionization states. These states lie above the ionization limit for an external electron and are due to transitions of the electrons of the inner shells of the atom. As a rule, the lifetime of an atom in an autoionization state is short (on the order of $10^{-12} \pm 10^{-13}$ sec), since it is determined mainly by the process of nonradiative transition into the ionization state. Autoionization states are of scientific interest for the understanding, identification, and interpretation of atomic spectra of multielectron atoms,[1] as well as of practical value when it comes to increase the cross section of ion photoionization by laser radiation.[2]

We report here the first observation of an autoionization state with an approximate lifetime 10^{-9} sec in the spectrum of a gadolinium atom.

2. The gadolinium atom were excited in the beam into states that lie above the ionization limit, in three steps, by radiation from pulsed tunable dye laser (emission line width $\Delta\nu = 1\ \text{cm}^{-1}$). The dye lasers were pumped simultaneously by a pulsed nitrogen laser (pulse power 350 kW, pulse repetition frequency 12 Hz). Figure 1 shows a simplified level scheme of the gadolinium atom. The dye laser of the first excitation stage, with $\lambda_1 \approx 5618$ Å took the gadolinium atoms from the ground $4f^7 5d\ 6s^2\ {}^9D_2^0$ state to the state $4f^7 5d\ 6s6p\ {}^9D_3^0$. The second laser with $\lambda_2 = 6351.7$ Å excited the atoms to the state $4f^7 5d\ 6s7s\ {}^9D_4^0$. The wavelength of the third laser was tunable in the range 6300–6100 Å, so that Rydberg states and states within 300 cm^{-1} above the ionization limit could be excited. The laser beam crossed the atomic beam in the region between electrodes, to which a rectangular voltage pulse was applied 20 nsec after the laser pulses. The produced ions passed through a slit in one of the electrodes and were registered again by a secondary electron multiplier (SEM). The signal from the SEM was stored and then fed to an *x-y* recorder. The other coordinate of the recorder was controlled by a signal proportional to the change of the frequency of the third-stage laser.

3. Figure 2a shows the dependence of the ion signal for a gadolinium atom on λ_3 when the wavelength is smoothly varicd in the range 6110–6240 Å. A detailed investi-

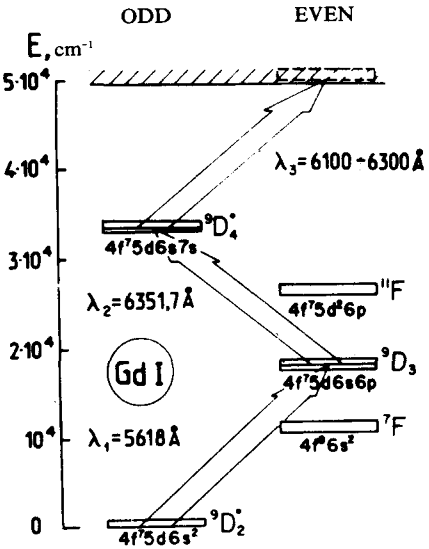

FIG. 1. Energy level scheme of godolinium atom, and the employed transitions.

gation of the behavior of this dependence in the region $\lambda_3 = 6220$–6225 Å made it possible to locate the ionization limit of the gadolinium atom. The value obtained for the ionization energy, $I = 495\ 98 \pm 5$ cm⁻¹, is in very good agreement with the data of Ref. 3. Attention is called to the intense autoionization resonance with $\overline{\lambda}_3 = 6133.5$ Å.

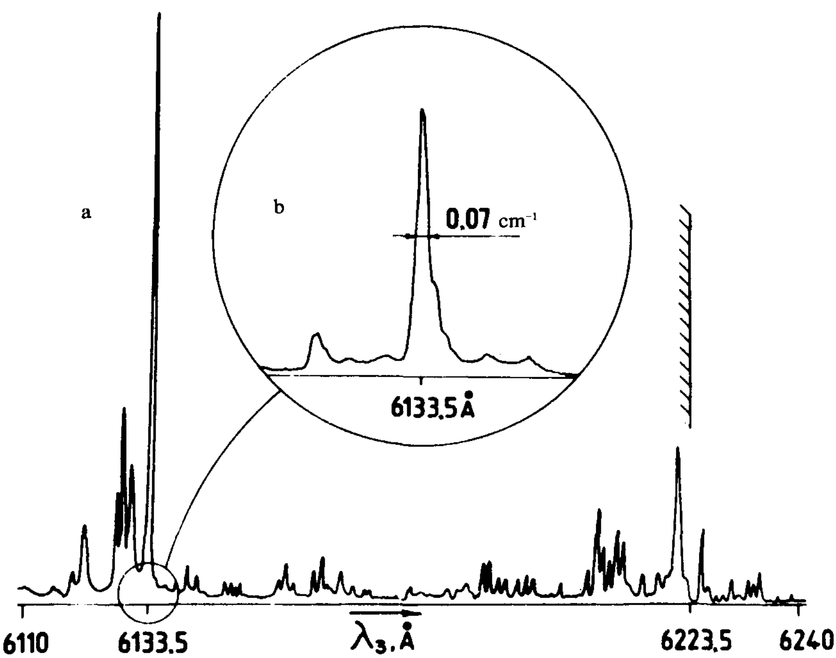

FIG. 2. a) Dependence of the ion signal on the wavelength of the third-stage laser, lasing line width $\Delta\nu_3 = 1$ cm⁻¹. b) The same dependence in the vicinity of $\overline{\lambda}_3 = 6133.5$ Å, lasing line width $\Delta\nu_3 = 0.03$ cm⁻¹.

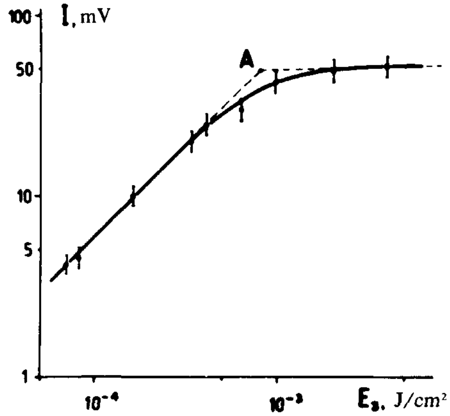

FIG. 3. Dependence of ion yield at the maximum of the autoionization resonance with $\overline{\lambda}_3 = 6133.5$ Å on the pulsed energy density of the third-stage laser, $\Delta\nu_3 = 0.03$ cm^{-1}.

Figure 2b shows this section of the spectrum with high resolution. The third-stage excitation was produced by a pressure-tuned dye laser with a lasing line width $\Delta\nu_3 = 0.03$ cm^{-1}. The width of the autoionization resonance at half-height is about 0.07 cm^{-1}, thus yielding an estimate $\tau \sim 0.5$ nsec for the lifetime of the state. Since the observed state is only 230 cm^{-1} away from the ionization limit, it decays most probably into a state consisting of an electron plus a gadolinium ion in the ground state. It appears that the selection rules forbid such a decay, and this leads to an increase of the lifetime of the state. This hindrance can be lifted, for example, in an electric field. Broadening of the autoionization resonance to 0.35 cm^{-1} was registered in a relatively weak electric field $\mathscr{E} = 100$ V/cm.

The cross section of this autoionization transition was measured by the method proposed in Ref. 4. Figure 3 shows the yield of the ions at the maximum of the resonance (Fig. 2b) against the energy density of the E_3 of the pulse of the third-stage laser. The point A is characterized by the condition $\sigma_{\rm aut}E_3 \approx 2h\nu_3$, whence we get the estimated cross section $\sigma_{\rm aut} \approx 8 \times 10^{-16}$ cm^2.

4. This experiment has shown that autoionization states with lifetimes on the order of 10^{-9} sec can exist in the spectrum of multielectron atoms. The cross section for the excitation of these states becomes comparable with the cross section for the excitation of bound high-lying states, and these long-lived autoionization states can be effectively used in the method of stepwise ionization of atoms by laser radiation to obtain a radical increase in the photoionization cross section of the atom.

[1]D.J. Bradley, P. Ewart, J.N. Nicholas, J.R.D. Shae, and D.G. Thompson, Phys. Rev. Lett. **31**, 263 (1973).
[2]V.S. Letokhov, V.I. Mishin, and A.A. Puretzky, Progress in Quantum Electronics **5**, part 3, 1939 (1977); V.S. Letokhov, V.I. Mishin, and A.A. Puretskiĭ, Khimiya plazmy (Plasma Chemistry), Atomizdat, 1977, No. 4, p. 3.
[3]E.F. Worden, R.W. Solars, J.A. Paisner, and J.G. Conway, J. Opt. Soc. Am. **68**, 52 (1978); W.C. Martin, R. Zalubas, and L. Hagan, Atomic Energy Levels–The Rare-Earth Elements, NS RDS–NBS 60, Washington, 1978.
[4]R.V. Ambartsumyan, V.M. Apatin, V.S. Letokhov, A.A. Makarov, V.I. Mishin, A.A. Puretskiĭ, and N.P. Furzikov, Zh. Eksp. Teor. Fiz. **70**, 1660 (1976) [Sov. Phys. JETP **43**, 866 (1976)].