Laser-spectroscopy measurement of the cross section of excitation transfer in a gas of like atoms

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The cross section for the transfer of excitation of monatomic europium vapor is determined experimentally. A change in the spectrum of the luminescence excited by the radiation of a narrow-dye laser, following excitation transfer by collisions, is revealed by the isotopic structure of the luminescence spectrum of europium.

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Excitation transfer in a gas consisting of one sort of atoms has by now been investigated in sufficient detail, and the cross sections of these processes can be calculated within the framework of the binary model with high degree of accuracy.[1] At present, however, there are no direct experimental data on the heavy-atom cross sections, knowledge of which is of independent interest for the physics of atomic collisions.

Processes of excitation transfer in a gas with a single sort of atoms manifest themselves at relatively high density of the colliding atoms. A direct experimental investigation by ordinary spectroscopic methods is hindered by the dragging of the radiation.

We propose here a method, free of these shortcomings, of directly observing and measuring the cross sections of processes of excitation transfer in a gas of like atoms. The method was experimentally realized for europium vapor.

For heavy atoms, a convenient object for the investigation of excitation-transfer processes is the transfer of excitation between individual isotopes in the atomic vapor. In this case, the conditions that the excitation transfer be resonant are satisfied, since the energy defect of the interacting atoms is negligibly small, and the atoms themselves are identical enough. At the same time, from the spectroscopic point of view the isotopes are well distinguishable, and this makes it possible to study independently the excitation transfer by determining the isotopic structure of the spectrum of the atoms.

The experiment was performed on metallic europium vapor. The necessary density of the atomic vapor was obtained by evaporating the metal from a molybdenum crucible into a vacuum chamber. The initial vacuum was 10^{-6} Torr and the crucible temperature was raised to 1200 °C, while the maximum europium vapor density was 10^{15} cm^{-3}. The vapor was exposed, directly at the opening of the crucible, to radiation of a narrow band ($\Delta\nu = 1.3\times10^{-3}$ cm^{-1}) cw laser based on rhodamine-6G. The laser power was 100 mW. The laser was used for selective excitation of one of the Eu isotopes on the transition $^8S_{7/2}$—$^6P_{7/2}$ ($\lambda = 5765$ Å). The luminescence accompanying this excitation was registered with a photomultiplier and an x–y recorder. The spectral analysis of

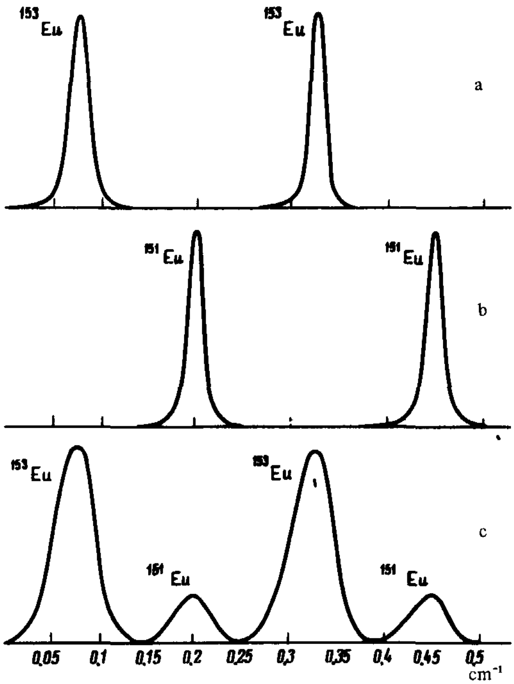

FIG. 1. Spectra of luminescence in ^{153}Eu and ^{151}Eu vapor: a—^{153}Eu, b—^{151}Eu, c—spectrum of luminescence produced in the transfer of excitation from ^{153}Eu to ^{151}Eu.

the luminescence was carried out by a scanned Fabry-Perot interferometer with a free spectral range that could be varied from 5 to 3.3×10^{-2} cm^{-1}, and with a sharpness 35.

At low atomic-vapor density we registered, using the dependence of the intensity on the wavelength of the exciting radiation, the isotopic structure of the chosen optical transition. The laser was then tuned to one of the isotopes and the luminescence spectrum of this isotope was registered with a scanning Fabry-Perot interferometer. As the atom-vapor density was increased, changes were observed in the luminescence spectrum, as manifested by a broadening and by the appearance of the luminescence line of the isotope that was not radiation-excited. Figures (1a)—(1c) show plots of the luminescence spectra both at low density of the Eu atoms, when the radiation excites the atoms ^{153}Eu or ^{151}Eu, and at high density, when the excited ^{153}Eu atoms transfer the excitation to the ^{151}Eu atoms. Since the onset of the luminescence line of the nonradiatively excited isotopes is due to the collision process of resonant excitation transfer, knowledge of the density of the atomic vapor in the irradiation region makes it possible to determine the cross section of this process.

Indeed, the experimentally measured ratio of the luminescence intensities of atoms A and B is equal to the ratio $a = N_A^*/N_B^*$ of the densities of the excited atoms. At the same time, it follows from the kinetic equations that describe, in the two-level approximation, the processes of radiative excitation of atoms A and of the collisional excitation transfer between the atoms A and B, under stationary conditions and assuming spontaneous decay of the excited states and an equal rate A_{21} for A and B, that the transfer cross section σ can be determined if we know the ratio $b = N_A/N_B$ of the concentrations of atoms A and B, and their combined density N:

$$\sigma = \frac{A_{21}}{a - b} \frac{1 + b}{N} \frac{1}{v},$$

where v is the average velocity of the atoms.

Measurements in the density range from 5×10^{12} to 5×10^{14} cm^{-3} yields for the cross section of the collision excitation transfer between the isotopes ^{153}Eu and ^{151}Eu a value equal 1.4×10^{-13} cm^2.

We note in conclusion that the method of direct observation of the excitation transfer process and of the measurements of its cross section can be used also to study the energy migration within the Doppler contour of the line.

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