Resonant scattering of low-energy electrons by mercury atoms

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An experimental setup is described for the measurement of elastic and inelastic scattering of electrons by atoms, and the first results are reported of an investigation of scattering of low-energy electrons (4.7–12 eV) in mercury vapor through an angle 90°. The energy dependences of the scattering cross sections reveal a resonance structure that correlates well with the structure on the optical spectral-line excitation functions. The spectra of the emitted electrons released as a result of the decay of the autoionization states at various bombarding-electron energies were also investigated, and a number of heretofore unobserved autoionization states of the mercury atom were found.

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INTRODUCTION

Much progress was made in recent years in the research on the resonant structure in the energy dependences of the cross sections for the scattering of electrons by atoms and molecules.[1] It was first observed in the form of sharp maxima and minima on the optical excitation functions of a number of spectral lines of mercury atoms.[2,3] The additional maxima were attributed by the authors to excitation of higher energy levels of the atom, followed by population of the initial level of the line via cascade transitions. It was shown in Ref. 4, however, that the primary cause of the fine structure on the optical excitation functions is not a cascade transitions but formation and decay of short-lived negative-ion states. Precision measurements of the total excitation cross sections have previously enabled one of us to separate and estimate the principal parameters of more than 15 resonances in the mercury atom.[4]

A large number of resonances was observed in experiments on the passage of an electron beam through mercury vapor (transmission experiments).[5] The energy dependences of the cross sections of elastic scattering of electrons by mercury atoms at various angles in the energy interval 4–6 eV were measured in Ref. 6. Four resonances were observed at 4.55, 4.71, 4.94, and 5.51 eV. The energy positions of the last three resonances are in good agreement with the data of Ref. 4, namely 4.70, 4.90, and 5.50 eV. A classification of the resonances observed in mercury is given in Refs. 7 and 8.

However, despite the apparent abundance of studies of electron scattering by mercury atoms, many aspects of elastic and inelastic scattering remain unknown; in particular, no study was made of resonances in the elastic channel in a wide energy interval, there are no data on the excitation and decay of low-lying autoionization states, and so on.

In the present paper we describe the experimental setup and present the first results of an investigation of elastic and inelastic scattering of low-lying electrons (4.7–12 eV) through an angle 90° in mercury vapor. In one experiment we studied, for the first time ever, the electron energy-loss spectra and their energy dependences, as well as the emission spectra of the electrons released as a result of autoionization.

EXPERIMENTAL SETUP

The experiments on the scattering of electrons by mercury atoms were performed by a volume method using a vapor-filled cell and differential evacuation of the collision chamber and of the electron-optical system. The principal units of the setup are a vacuum chamber emptied by a VA-05-4 vacuum unit, an electron spectrometer, a system for the registration of the current of the scattered (emitted) electrons, and the spectrometer control block.

The electron spectrometer (see Fig. 1) consists of an electron gun, a vapor-filled cell, and a 127° electrostatic energy analyzer for electrons scattered through 90°. The electron gun, with indirectly heated cathode, contains an electrostatic lens[9] (electrodes A_3–A_5 in Fig. 1) to shape an electron beam of specified energy and geometry. The system is provided with a small conical electrode A_6 ahead of the collision chamber, intended for additional collimation of the beam. The beam geometry was monitored by measuring the current to two concentric collectors (A_7, F). The collision zone was 6 mm long. The vapor pressure in the cell did not exceed 5 $\times 10^{-3}$ Torr and was determined by the temperature of a mercury-filled container located outside the vacuum

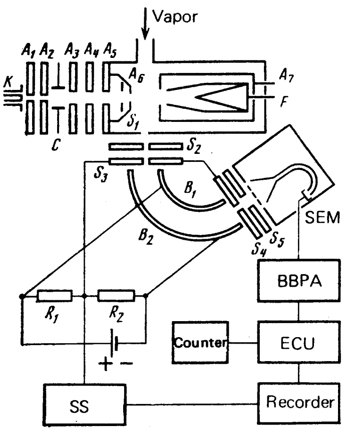

FIG. 1. Block diagram of experimental setup (see the text).

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chamber. The primary electron beam current in the collision zone was ~5 × 10^{-6} A with an FWHM energy spread 0.3–0.4 eV.

An energy analysis of the electrons scattered through 90° relative to the incident beam was effected with a 127° cylindrical electrostatic Hughes–Rojansky analyzer of construction similar to that described in Ref. 10. The energy resolution of the analyzer in our experiments was 0.04–0.1 eV. To detect the current of the scattered electrons we used a VÉU-6 channel secondary-electron multiplier (SEM) operating in the single-pulse counting regime. The SEM pulses amplified by a broadband preamplifier (BBPA) were fed to the input of the recording system, comprising an ÉVU 1-1 electronic computing unit (ECU) that could feed the information to a counter (frequency meter ch3-38) and an x-y recorder (PDS-021).

To record the energy-loss and emitted-electron spectra in the constant resolution regime[11] a control voltage was applied to the input electrode of the energy analyzer S_3 from the scanning source (SS). The latter was either a G6-15 special-waveform signal generator or a VK2-21 calibrated-voltage source. The maximum SEM pulse count did not exceed 10^5 sec^{-1}.

MEASUREMENT RESULTS AND THEIR DISCUSSION

The spectrometer resolution turned out to be sufficient to separate all the lower energy levels of the mercury atom. We discuss below the results of our measurements of the energy dependences of the intensity of some of the loss-spectrum lines (Figs. 2–4), which duplicate the energy dependences of the differential cross sections (DS) for scattering through 90°.

The energy dependences of the DS for elastic scattering and excitation of resonance levels in the energy region 4.7–6.5 eV are shown in Fig. 2. The same figure shows the data of Refs. 6 and 4 on elastic scattering and the total excitation cross section Q of the levels $6^3P_{0,1}$ and 6^3P_2. It is seen that good agreement is observed in the behavior of the elastic-scattering curves, and some deviations are attributed to the different energy resolutions of the instruments. However, at an energy ~5.2 eV we have observed some deviation of the elastic-scattering cross section from monotonicity. This is apparently due to the appearance of a resonance caused by the decay of the $^2D_{3/2}$ state of the negative Hg$^-$ ion,[8] which was observed also on the optical excitation function (EF) of the mercury resonance line λ 2537 Å.[10] We note that the resonance at 5.50 eV served as a reference for the absolute energy scale.

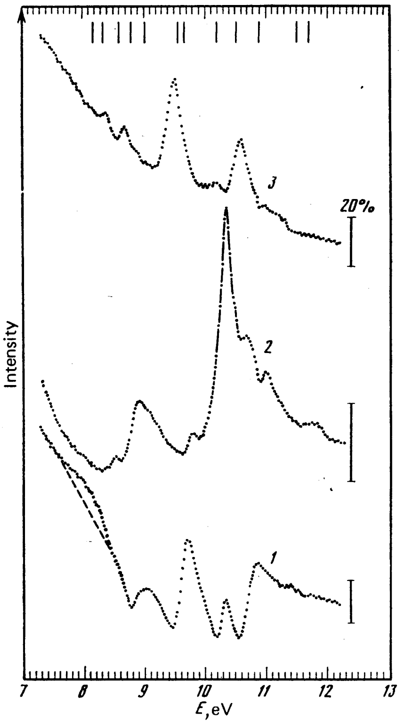

FIG. 3. Energy dependences of the DS of elastic scattering (curve 1), of the DS of the excitation of $6^3P_{0,1}$ level (curve 2) and of the 6^3P_2 level (curve 3) of mercury. The vertical bars mark the positions of the resonances in the optical channel.[4]

Two resonances were previously[4] separated on the EF of the mercury line λ 2537 Å (6^1S_0–6^3P_1) and in the total excitation cross section of the 6^3P_1 level in this energy region, namely, at energies 4.90 and 5.50 eV, with respective widths ~0.1 and 0.25 eV. As seen from Fig. 2, at these energies the resonances manifest themselves

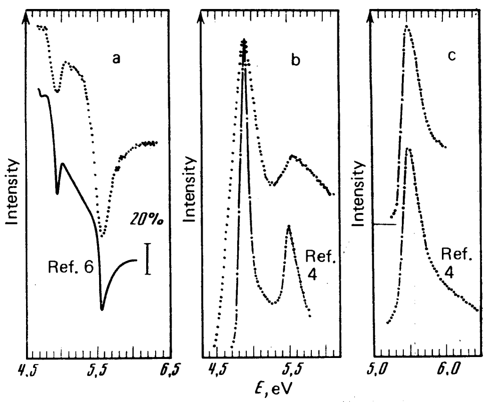

FIG. 2. Energy dependence of the DS: a—of elastic scattering, b—of excitation of $6^3P_{0,1}$ level, and c—of excitation of 6^3P_2 level of mercury.

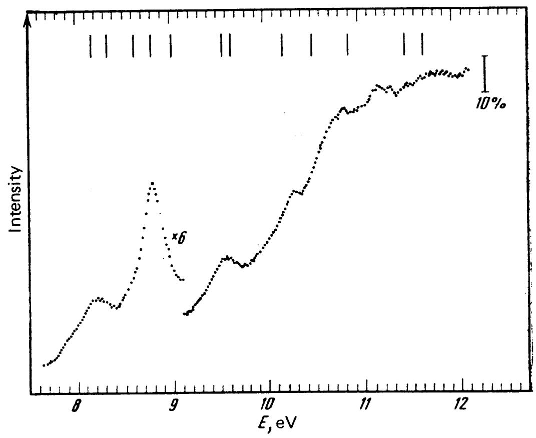

FIG. 4. Energy dependence of the DS of the excitation of the 6^1P_1 level of mercury.

in the elastic-scattering cross section (in the form of deep minima) and in the energy dependence of the DS for the excitation of the $6^3P_{0,1}$ level (in the form of sharp maxima). The resonance at 5.50 eV is seen also in the DS for excitation of the 6^3P_2 level. Thus, in this energy interval, complete correlation of the singularities in the total (optical and inelastic channels) and the differential (elastic and inelastic channels) cross sections for scattering is observed, and the earlier[4] assumption that the negative ions decay via many channels is indeed true.

The monotonically decreasing curve of the elastic-scattering DS (Fig. 3, curve 1) shows at 8.2 eV a maximum of width ~0.6 eV. This is due to the appearance of the resonance ($E = 8.18$ eV) previously observed in the optical channel in the excitation of the 7^3S_1 level. More clearly pronounced is the resonance at $E = 8.80$ eV. The center of the resonance is located at the minimum of the DS. The minimum of the DS at $E = 9.6$ eV agrees well with the position of the 9.56 eV resonance in the optical channel; the latter was observed on the EF of eight spectral lines. The resonance at 10.20 eV is represented as an alternation of a minimum and a maximum. A broad maximum is observed at 10.9 eV and is evidence of appearance of the resonance previously observed on the EF of a large number of spectral lines.

We compare now the structure in the DS for the excitation of the $6^3P_{0,1}$ level, on the one hand, with the resonances in the total cross section for the excitation of this level.[4,12] The narrow resonance at 8.34 eV appears in the DS as an alternation of a minimum and a maximum (see Fig. 3, curve 2). A similar picture is observed in the case of the resonances at 8.8 and 9.6 eV. The high maximum at 10.34 eV is observed also on the EF of the λ 2537 Å line. The resonance near 10.9 eV manifests itself in the DS as an alternation of a maximum and a minimum. A maximum in the inelastic channel corresponds also to a maximum in the elastic channel. As seen from Fig. 3, the singularities typical of the EF of the λ 2537 Å line are preserved in the energy dependence of the DS. Thus, the main causes of the structure observed on the optical EF of the spectral lines are not cascade transitions but formation and decay of negative-ion states.

A resonance structure can also be traced on the energy dependence of the DS for the excitation of the 6^1P_1 level (Fig. 4). One of us has previously noted[13] a correlation between the extrema in the excitation of the $^3S_1$ and $^1P_1$ levels of the Cd and Mg atoms.[13] A corresponding correlation of the structure at 8.5 eV was expected also in the cross sections for the excitation of the pair of mercury spectral lines λ 5461 Å ($6^3P_2-7^3S_1$) and λ 1850 Å ($6^1S_0-6^1P_1$). The EF of a single resonance line, however, could not be investigated because of the low sensitivity of the radiation detector in the vacuum ultraviolet. In this connection, particular interest is attached to the study of the energy dependence of the DS for the excitation of the 6^1P_1 level. As seen from Fig. 4, near the electron energy 8.6 eV there is alternation of a minimum and a maximum, thus attesting to the process of a resonance. It can therefore be assumed that in this energy region a correlation of the singularities should also be observed on the EF of the indicated lines.

We consider now the structure of the cross sections beyond the ionization threshold. Maxima of the fine structure were observed also on the optical EF of a large number of spectral lines of mercury beyond the ionization threshold.[4] As seen from the presented results, the differential cross sections also have a structure in this energy region. Thus, for example, there is a clearly pronounced resonance at 10.9 eV in the elastic channel (Fig. 3, curve 1). Weaker singularities are observed also at energies 11.1, 11.3, and 11.7 eV. In the DS for the excitation of all the levels measured by us one can note also extrema lying beyond the ionization threshold. In Ref. 4, the extrema at 10.90, 11.56, and 11.72 eV on the optical EF were classified as Feshbach resonances. It was assumed that the originating states of these resonances are the mixed levels $5d^{10}6p^2\ ^3P_0$ and $^3P_1$, and the Beutler levels $5d^96s^26p\ ^3P_1$ and $^3D_1$. However, besides the resonances indicated above, this energy region contains a considerable number of maxima that do not lend themselves to graphic analysis because of the strong overlap of neighboring resonances. The question of the origin of resonances has therefore remained open.

It is known[14] that besides the decay of the short-lived states of negative ions, the excitation cross section can be significantly influenced by the so-called effect of post-collision interaction of the emitted and scattered electrons. This effect leads to a shift of the structure on the EF lines within one spectral series or to a change in the energy of the emitted electrons near the autoionization threshold of the states of the atom.

To clarify the causes of the appearance of the structure in the scattering cross section beyond the ionization threshold, we investigated the spectra of the electrons released in the decay of autoionization states. Figure 5 shows a spectrum section corresponding to the

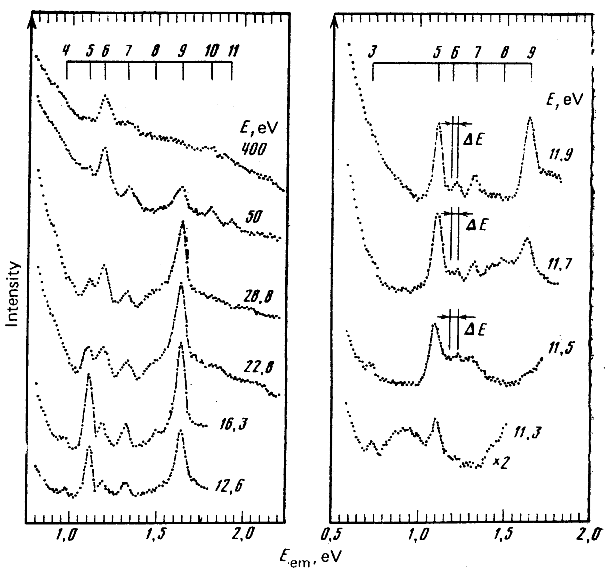

FIG. 5. Spectra of the electrons emitted as a result of the decay of autoionization states at various energies of the incident electrons E.

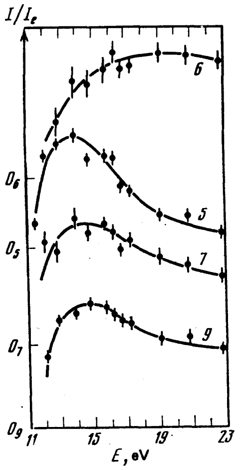

FIG. 6. Energy dependences of the DS of the excitation of four autoionization levels (the line intensities I are normalized to the beam current I_e).

decay of the autoionization states lying near the ionization threshold of mercury at various incident electron-beam energies.[1)]

The excitation energies E_a of the observed autoionization levels of the mercury atom are listed in the table. It gives also the values of the excitation energies of the states 1 and 2, observed by the method reported in Ref. 16, as well as a comparison with the theoretical-calculation data.[17] It is seen that besides the levels previously observed in experiment,[18] in this energy interval there is observed the decay of nine more autoionization states, seven of which are most clearly pronounced at low energies. The line widths indicate that these levels are apparently long-lived. The measured energy dependences of the DS for the excitation of a number of lines in the spectrum (Fig. 6) confirmed this assumption. The intensities of lines 5, 7, and 9, for example, have rather distinct maxima near the threshold, while the optically allowed line 6 is most effectively excited at high energies.

When the electron-beam energy decreases to the threshold of the autoionization states, a noticeable shift of the maximum of line 6 ($6p^3D_1$ state) is observed towards higher energies thus indicating the presence of a post-collision interaction. The registered maximum shift is 0.05 eV, which is much higher than the accuracy with which the energies of the emitted electrons are determined (±0.01 eV). For a more unequivocal determination of the causes of this effect it is obviously necessary to investigate in detail the optical EF of the spectral lines corresponding to transitions from the high-lying energy levels of the mercury atom.

An analysis of the structure on the EF and of the spectra of the emitted electrons shows that in most cases the singularities, both in the energy dependences of the scattering of the DS and on the optical EF, lie below the energy of the corresponding lines in the spectrum of the emitted electrons. It can therefore be concluded that the structure in the cross sections of the elastic scattering and of the excitation of the lower levels of mer-

TABLE I.

Line No.	E_a, eV	E_{opt}, eV	State	J	Theory[17]	[18] **
0*	10,61	0,18		2	0,19	
1	10.79	0,36				
2	11.07	0,64	$6p\,^3P_1$	1	0.57	0.57
3	11.17	0,74		0	0.76	
4	11.40	0,97				
5	11.54	1.11		3	1.11	
6	11.62	1.19	$6p\,^3D_1$	1	1,18	1,19
7	11.74	1,31		2	1.34	
8	11.92	1,49				
9	12.07	1.64				
10	12.25	1,82	$2e$			
11	12,36	1.93	$2e$			

*Determined from the energy loss 10.61 eV.
**Experiments on UV absorption.

cury is due to decay of short-lived states of the negative ion Hg^-. The large number of low-lying autoionization states that can serve as the initiating states of the resonances, and the many channels of the decay of the negative ions, cause the complicated character of the cross sections for electron scattering by mercury atoms at energies exceeding the ionization threshold.

The reported investigations provide a new experimental confirmation of the substantial role of the short-lived states of negative ions in autoionization states when low-energy electrons are scattered by mercury atoms, and of the large number of channels of their decay into an electron and an atom in the excited states. They are the first step in a study of scattering of low-energy electrons by electron spectroscopy.

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Narrow nonlinear nonresonances in a three-level system

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We investigate the possibility of transferring a narrow line-shape singularity from one of the transitions of a three-level gas system to an adjacent transition via coupling through a common level. The density matrix is determined at arbitrary saturation with respect to both resonant fields, neglecting higher spatial harmonics. Explicit expressions are obtained for the nonlinear susceptibilities at small saturation in the Doppler limit. The contrast and width of the transferred singularity (formed by an absorbing cell) in two-frequency generation on standing waves are determined. The system He–Ne/CH_4 is considered by way of example.

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1. INTRODUCTION

It is known[1,2] that the use of narrow nonlinear resonances which arise when a low-pressure absorbing cell is placed inside the resonator, makes it possible to form on the broad ($\sim ku_0$) Doppler contour an amplification peak (an inverted Lamb dip) with a width determined by the homogeneous broadening γ of the absorption line. A classical system of this type is the amplifying medium He–Ne with a methane absorbing cell ($\lambda = 3.39$ μm).[2] An attempt can be made to get around the difficulties of producing such a system for shorter wavelengths, for example in the optical band, which are connected with the absence of such narrow absorption lines, by using the idea[3] of transferring the narrow Lamb detail to an adjacent high-frequency transition in a three-level active medium. Owing to the presence of the common level, the adjacent transitions compete, therefore a narrow resonance on one of the transitions should appear also on the adjacent one.

This possibility is theoretically investigated in the present paper. Expressions are obtained for the density matrix, describing the gas three-level system in a resonant field,[1,4] in the case of waves with arbitrary amplitudes and standing-wave coefficient. The nonlinear susceptibilities averaged over the velocities are obtained for small saturation parameters. In the Doppler limit $ku_0 \gg \gamma$, expressions are obtained for the line shape, and they are analytically investigated for small detunings from resonance. It is shown that the appearance of a narrow (less than the homogeneous width) singularity on one of the transitions can lead to the appearance of a singularity on an adjacent transition. It is useful to employ two-frequency lasing for this purpose.[5-10] The contrast and width of the "transferred" singularity in two-frequency lasing on the standing waves are determined. It is shown that for the system He–Ne/CH_4 the transferred singularity is a dip rather than a peak, in contrast to the assumption made in Ref. 3.

2. DENSITY MATRIX IN RESONANT FIELDS

The equation of motion for the single-particle density matrix, which is an operator in the internal variables and a classical distribution function in the coordinates of the center of gravity of the particles, takes the form[1,11]

$$\left(\frac{\partial}{\partial t}+\mathrm{v}\nabla\right)=-\frac{i}{\hbar}[\hat{H},\hat{\rho}]-\hat{\gamma}(\hat{\rho}-\hat{\rho}_0), \tag{2.1}$$

where $\hat{H}=\hat{H}_0+\hat{V}(z,t)$ is the Hamiltonian of the particle; $\hat{\gamma}$ is the matrix of the relaxation constants,

$$(\hat{\gamma}\hat{\rho})_{nm}=\gamma_{nm}\rho_{nm},$$

$\hat{\rho}_0$ is the stationary density matrix in the absence of a field. In the presence of resonant fields, the matrix elements of the perturbation are equal to

$$V_{nm}(z,t)=\sum_{\alpha=\pm1}V_{nm}{}^{\alpha}\exp\{i(\alpha k_{nm}z-\Omega_{nm}t)\}+\text{c.c.}, \tag{2.2}$$

where $V^{\alpha}_{nm}=-\mathbf{d}_{nm}\mathbf{E}^{\alpha}_{nm}$, $\mathbf{d}_{nm}$ is the dipole moment, $\mathbf{E}^{\alpha}_{nm}$ is the amplitude of the resonant field on the $n\to m$ transition, the superior index $\alpha=\pm1$ distinguishes between waves traveling in opposite directions; Ω_{nm} are their frequencies, and k_{nm} are the wave numbers, with $\Omega_{nm}=-\Omega_{mn}$, $k_{nm}=-k_{mn}$. As follows from the structure of (2.1), stationary solutions in the resonance approximation with allowance for the lower spatial harmon-

