Experimental Determination of Auger-Decay Amplitudes from the Angular Correlations in Auger Cascade Following the $2p \rightarrow 4s$ Photoexcitation in Ar

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We have determined the ratio of the s and d Auger-decay amplitudes and their phase difference for the resonant Auger transition $2p_{3/2}^{-1}4s \rightarrow 3s^{-1}3p^{-1}4s(^2P)$ from the measurement of the angular correlation between the resonant Auger emission and the subsequent second-step Auger emission, as well as the angular distribution measurements for these two electron emissions. The analysis of the experimental data is based on the nonrelativistic *LSJ* coupling approximation.

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The experiment for determining all of the dynamical parameters (amplitudes) characterizing a particular atomic process, often called a *complete* or *perfect* experiment, has fundamental importance in quantum physics. Being determined from the experiment, the amplitudes permit one to predict *all* other characteristics of the process even though some of them are impossible to measure with modern facilities. The complete experiment provides also the ultimate test of any theoretical descriptions of the process. In spite of their importance complete experiments are still rare since they usually demand very sophisticated techniques such as spin-polarization measurements, coincidence methods, target and/or projectile polarization, etc. For atomic photoionization, several approaches to complete experiments have been suggested and realized in the last two decades. Heinzmann carried out the first complete measurement by measuring the angular distributions and the spin polarizations of photoelectrons [1]. Hausmann *et al.* combined the angular distribution measurements for photoelectrons and for Auger electrons from the photoion [2] to achieve complete characterization of the photoemission process. Similarly Jiménez-Mier *et al.* used the angular distributions of photoelectrons and fluorescent photons [3]. Kämmerling and Schmidt made coincidence measurements of the angular correlations between photoelectrons and Auger electrons [4]. Also coincidence measurements, but in their case for photoelectrons and polarized fluorescence photons, were used by Beyer *et al.* [5]. Finally, very recently the angular distributions of photoelectrons from polarized atoms have been used [6,7] to realize the complete experiment in photoionization.

A similar goal of complete characterization may be pursued for the Auger-decay process. The possibility of performing a complete experiment in Auger spectroscopy was first considered theoretically by Kabachnik and Sazhina [8]. It was shown that the complete experiment is possible and several ways of its realization were discussed. However, due to low intensity of the Auger lines and their small anisotropy and spin polarization, until quite recently there were no reported attempts to obtain experimentally all of the Auger-decay amplitudes. In this Letter we report one of the first attempts of an *almost* complete experiment for Auger decay: the experimental determination of the ratios of the amplitudes and their phase difference with the use of angular correlation measurements in an Auger cascade. A similar kind of complete experiment was carried out recently by West *et al.* [9] and Ueda *et al.* [10] for autoionization of photoexcited Sr atoms, by measuring the angular correlation between the autoionization electron and the subsequent fluorescent photon. Grum-Grzhimailo *et al.* [11] also discussed the complete experiment for Auger decay in a different approach based on measurements of the intensity ratio and the angular distributions of the Auger electrons.

The tunability of synchrotron radiation allows one not only to ionize an electron of a specific innershell but also to excite it to a specific orbital. The innershell-excited atomic state decays *via* a resonant Auger transition. A particularly interesting case is when the ion formed in the resonant Auger decay can decay further by emitting one more electron. In this case, one can gain some dynamical information on the decay process of the innershell-excited state, as well as spectroscopic information for the states involved, from the angular correlation measurement between the resonant Auger and the second-step Auger electrons as well as the angular distribution measurements for these two electrons, and indeed one can design a *complete* experiment for the decay process [8,12,13].

As a specific example of such cascade processes, we consider here the following cascade in Ar photoexcited to the $2p_{3/2}^{-1}4s$ state by linearly polarized light:

$$\mathrm{Ar}(^1S_0) + h\nu \rightarrow \mathrm{Ar}^*[2p^5 3s^2 3p^6(^2P_{3/2})4s, J_0 = 1], \tag{1}$$

$$\rightarrow \mathrm{Ar}^{*+}[2p^6 3s 3p^5(^1P_1)4s\,^2P_{J_1}] + e^-(l_1 j_1), \tag{2}$$

$$\rightarrow \mathrm{Ar}^{++}(2p^6 3s^2 3p^4\,^3P_{J_2}) + e^-(l_2 j_2), \tag{3}$$

where $J_1 = 1/2, 3/2$ and $J_2 = 0, 1, 2$. The corresponding Auger lines at an energy ~194 eV [transition (2)] and ~7 eV [transition (3)] were observed in an electron-electron coincidence experiment [14]. The configuration of the intermediate state, $3s^{-1}3p^{-1}4s$, in (2) should be considered only as a label; this state is a mixture of this configuration with $3p^{-3}3d4s$ as noted in the theoretical investigations of Refs. [14–16].

The innershell-excited state formed by photoabsorption (1) of linearly polarized light is aligned along the light polarization axis. Because of this alignment, the resonant Auger decay (2) is anisotropic. Its anisotropy depends on the Auger-decay amplitudes (see, for example, [17]). Moreover, in resonant Auger decay, the alignment is partly transferred to the intermediate ($^2P_{3/2}$) state and the value of the alignment transfer parameter is determined again by the resonant Auger-decay amplitudes [12,13]. The second-step Auger decay in the cascade (3) is also anisotropic and its anisotropy reveals the alignment of the intermediate state. The angular correlation between the two emitted electrons, which can be measured in a coincidence experiment, provides additional parameters which are related to the Auger-decay amplitudes of both transitions [13], in complete analogy with the coincidence measurements of the photoelectron and the Auger electron [18] or the photoelectron and the fluorescent photon [19].

In this Letter, we report measurements for the angular correlation function between the resonant Auger electron emission (2) and the second-step Auger electron emission (3) in an Auger cascade, as well as measurements of the angular distribution of the resonant Auger electrons. We repeated also the angular distribution measurement of the second-step Auger emission (3) in order to improve the preliminary results given in [20]. Combining these results we obtain the experimental values of the resonant Auger-decay amplitudes and their phase difference, thus realizing the complete experiment.

The measurements were carried out on the 24-m spherical grating monochromator installed in the BL-16B undulator beam line at the Photon Factory [21], using apparatus described elsewhere [22]. The photon energy was tuned to the Ar $2p_{3/2} \rightarrow 4s$ excitation at 244.4 eV and the photon band pass was ~0.4 eV. The incident light was focused onto the interaction region, where light was merged with an effusive gas beam, ejected from an axial cell through eight straight needles of 0.5 mm in inner diameter. The ambient pressure in the experimental chamber was typically 4×10^{-5} Torr during the measurement and was isolated from the beam line by an Al filter of 1000 Å thickness.

In the measurement of the angular correlation between the resonant Auger emission (2) and the second-step Auger emission (3) in the plane perpendicular to the photon beam, we used two identical 150° spherical sector electron spectrometers with a mean radius of 80 mm (see inset in Fig. 1). The first spectrometer was mounted on a turntable, whose axis of rotation was aligned to coincide with the incident light beam, and detected the resonant Auger electrons at kinetic energy of ~194 eV ejected in the first decay (2). The second spectrometer was set in such a way that it detected Auger electrons at a kinetic energy of ~7 eV ejected in the second decay (3), perpendicular to the linear polarization axis of the incident light. The energy resolution was set to 1.5 eV for the first analyzer and 0.2 eV for the second analyzer. The typical electron counting rates were 400 and 300 cps for the first and second analyzers, respectively.

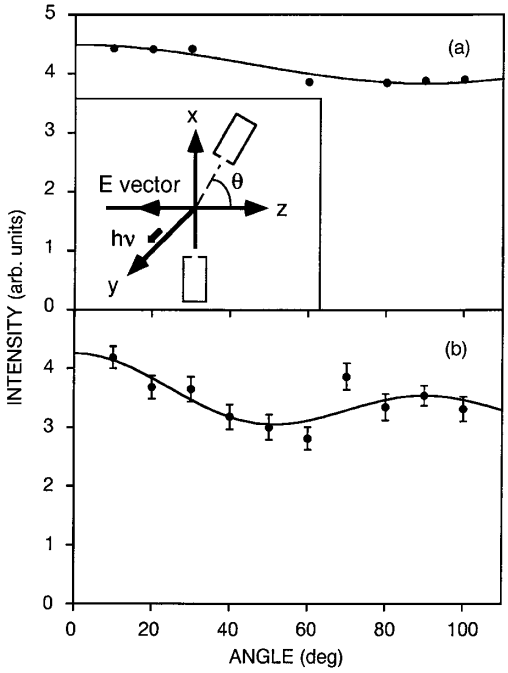

FIG. 1. Angular distributions for the resonant Auger electrons ejected in the first-step decay of the Ar $2p_{3/2} \rightarrow 4s$ excitation; (a) without detecting the second-step Auger electrons and (b) detecting in coincidence the second-step Auger electrons in the direction $\theta = 270°$. The solid lines in (a) and (b) correspond to the result of the fit using Eqs. (4) and (5), respectively. In the inset the kinematics of the experiment is shown.

The coincidence rate between these two electrons was measured as a function of the detection angle of the resonant Auger electrons relative to the linear polarization axis of the incident light. The false coincidence rate was in general significantly smaller (typically $\leq 10\%$) than the true coincidence rate, and only the true coincidence rate, obtained by subtraction of the false coincidence background, was used in the analysis. To remove the effect of the fluctuation of the coincidence counting rates as a function of time we normalized the coincidence counts by the electron counts detected by the fixed analyzer.

In addition, we measured the angular distributions for the resonant Auger electrons, by counting the electrons passing through the analyzer as a function of the electron detection angle relative to the polarization axis of the incident light. From the measurement for the Ar $3s$ photoelectron line, for which we expect β to be 2, the degree of

linear polarization $P_{\rm lin}$ of the incident light turned out to be $100^{+0}_{-3}\%$ in the horizontal plane.

Figure 1(a) shows the results for the angular distribution of the resonant Auger electrons emitted in the first decay (2). The electron counts were plotted as a function of the angle relative to the linear polarization axis of the incident light. The angular distribution of the resonant Auger electrons can be described by a well-known formula (the z axis is along the light polarization direction):

$$I(\theta) = I_0[1 + \beta^{(1)} P_2(\cos\theta)], \tag{4}$$

where $P_2(x)$ is a second-order Legendre polynomial, I_0 is a constant factor proportional to the cross section, and $\beta^{(1)}$ is the asymmetry parameter for the Auger emission. We have obtained the $\beta^{(1)}$ value through a least-squares fit of Eq. (4) to the data points in Fig. 1(a): the resulting value is 0.09 ± 0.05.

Figure 1(b) shows the angular distribution of the resonant Auger electron (2), recorded in coincidence with the second-step Auger electron (3) which is detected perpendicular to the photon polarization direction ($\theta = 270°$). It is clear that the anisotropy of the resonant Auger electrons is enhanced by detecting them in coincidence with the second-step Auger electrons. Also it is clear that the angular distribution cannot be described any more by Eq. (4). Instead it can be fitted to the expression:

$$I(\theta) = A_0 + A_2 \cos 2\theta + A_4 \cos 4\theta\,, \tag{5}$$

as we will discuss later. We have obtained the ratios A_2/A_0 and A_4/A_0 through a least-squares fit of Eq. (5) to the data points in Fig. 1(b): the resulting values are $A_2/A_0 = 0.10 \pm 0.05$ and $A_4/A_0 = 0.12 \pm 0.05$.

We repeated also the angular distribution measurement of the second-step Auger emission (3) and improved the $\beta^{(2)}$ value given in our preliminary report [20]: the resulting value is $\beta^{(2)} = -0.44 \pm 0.05$.

Now we discuss how the parameters $\beta^{(i)}$ and A_i/A_0 are related to the resonant Auger amplitudes and their phase difference. In a general (relativistic) case, according to the angular momentum and parity selection rules, the first-step decay is described by two (for the $^2P_{1/2}$ final state) and by three (for the $^2P_{3/2}$ state) complex partial amplitudes, $M_j^{J_1}$, corresponding to the s and $d_{3/2,5/2}$ partial waves of the ejected electron. However, in the nonrelativistic approximation, with some additional model assumptions discussed below the number of independent amplitudes describing the decay can be considerably reduced. The same is valid for the second-step Auger decay.

The resonant state $2p_{3/2}^{-1}4s$, i.e., the initial state for the first Auger decay, is aligned along the photon polarization (z axis). Using the conventional two-step ansatz, the angular distribution parameter $\beta^{(1)}$ for the resonant Auger electrons can be expressed as a product of two terms, the alignment parameter $\mathcal{A}_{20}$ of the initial state and the intrinsic anisotropy parameter α_2^{Σ} for the Auger decay [23]:

$$\beta^{(1)} = \mathcal{A}_{20}\alpha_2^{\Sigma}\,. \tag{6}$$

The superscript Σ is added to indicate that $\beta^{(1)}$ is the average asymmetry parameter for two experimentally unresolved transitions to the $^2P_{1/2,3/2}$ intermediate states of Eq. (2). In fact, they almost completely overlap, and in the following we ignore the energy splitting of the two states. The alignment parameter $\mathcal{A}_{20} = -\sqrt{2}$ for a state excited by completely linearly polarized light from the ground state of a closed-shell atom. In order to evaluate α_2^{Σ}, we assume that in transition (2) the intermediate state is well described within the *LSJ* approximation, i.e., the total orbital momenta L and total spin S are good quantum numbers. Note that the *LSJ* approximation does not exclude strong configuration mixing, which is important in the proper description of this state. Besides the *LSJ* approximation for the intermediate state, we assume that the spin-orbit interaction in the continuum can be neglected. Then all of the five amplitudes $M_j^{J_1}$ can be expressed in terms of two Auger amplitudes M_s and M_d for the s and d waves and their phase difference Δ. By expanding the wave function of the $2p_{3/2}^{-1}4s$ initial state in terms of *LSJ* functions and transforming the Auger matrix elements to the *LSJ* coupling scheme (for details, see [23]) we obtain the expression for the angular distribution parameter $\beta^{(1)}$ for the resonant Auger electrons in terms of three parameters M_s, M_d, and Δ:

$$\beta^{(1)} = \frac{M_d^2 - 2\sqrt{2}\,M_s M_d \cos\Delta}{2(M_s^2 + M_d^2)}\,. \tag{7}$$

Using the same approximations as those employed for the first-step Auger decay, we obtain an expression for the angular distribution parameter $\beta^{(2)}$ of the second-step Auger electrons (assuming the first-step electron is not detected in coincidence). Here the interference of the two overlapping intermediate states should be taken into account, as we have shown previously [20]. To do that, we use the general expressions for the angular distribution of Auger decay which include overlapping initial states [24]. The alignment transferred to the intermediate state is also determined by the first-step Auger amplitudes M_s and M_d. We note also that only the p wave contributes to the second decay $^2P_{1/2,3/2} \rightarrow {}^3P_{0,1,2}$ [see Eq. (3)], within the nonrelativistic *LSJ* approximation. Using the above approximations, calculating the alignment transfer as outlined by Ueda *et al.* [20], and summing over the unresolved states of the final multiplet ($^3P_{0,1,2}$), we obtain an expression for $\beta^{(2)}$ for the second-step Auger electrons:

$$\beta^{(2)} = -\frac{M_s^2 + 0.1\,M_d^2}{2(M_s^2 + M_d^2)}\,. \tag{8}$$

(Note that in Ref. [20] the minor d-wave contribution was neglected.)

The angular correlation between the two consecutively emitted electrons can be obtained using the approach described in Ref. [13] generalized to account for the overlapping intermediate states. It has the form of Eq. (5) with coefficients A_i depending on the Auger-decay matrix elements. Using the nonrelativistic *LSJ* approximation for both first-step and second-step Auger transitions, we eventually express the A_i coefficients in terms of the same parameters M_s, M_d, and Δ. We obtain

$$A_2/A_0 = \frac{48M_d^2 - 96\sqrt{2}\,M_s M_d \cos\Delta}{80M_s^2 - 16\sqrt{2}\,M_s M_d \cos\Delta + 61M_d^2}, \quad (9)$$

$$A_4/A_0 = \frac{27M_d^2}{80M_s^2 - 16\sqrt{2}\,M_s M_d \cos\Delta + 61M_d^2}. \quad (10)$$

Thus, within our model, we can express all the anisotropy coefficients for both noncoincidence and coincidence measurements in terms of only two independent parameters: the ratio of the decay amplitudes M_d/M_s and the phase difference ($\cos\Delta$). To determine these values from the experiment it is sufficient to measure only two observables. For example, one can use the results of the coincidence experiment only: this variant is analogous to the complete experiment in photoionization made by Kämmerling and Schmidt [4]. Alternatively one can use the values $\beta^{(1)}$ and $\beta^{(2)}$ as obtained from the noncoincidence measurements: this variant is analogous to the complete experiment in photoionization made by Hausmann *et al.* [2]. The most reliable values of M_d/M_s and $\cos\Delta$, however, can be obtained from the least-squares method treating M_d/M_s and $\cos\Delta$ as fitting parameters and four values $\beta^{(1)}$, $\beta^{(2)}$, A_2/A_0, and A_4/A_0 as data points to be fitted using Eqs. (7)–(10). The values thus obtained are $M_d/M_s = 0.52 \pm 0.15$ and $\cos\Delta = 0.01 \pm 0.03$.

In conclusion, we have demonstrated the first *complete* experiment for resonant Auger decay using the angular correlation measurement between the resonant Auger electron and the second-step Auger electron as well as the angular distribution measurements for the resonant Auger electron and the second-step Auger electron. The *completeness* however relies on the model one employs. In our case, we employed the nonrelativistic *LSJ* coupling approximation. If this approximation fails, the simple relations (7)–(10) are not valid and more parameters are necessary to describe the observed angular correlation coefficients. In this case complementary measurements are necessary (for example, the spin polarization of Auger electrons) for the experiment to be complete. It would be desirable to compare the present results with a first-principles theoretical calculation without the approximations employed here and such attempts are under current consideration.

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