The prominent series of Beutler-Fano shaped autoionized lines converges closely to 256 Å (48.5 eV) and most probably corresponds to the transitions $2s^22p^6\,{}^1S_0$-$2s2p^6np\,{}^1P_1{}^\circ$. Nine members of this series are observable on the original plates.

Many other discrete anomalies in the absorption spectrum of Ne can be seen in Fig. 1, and they differ considerably in character. The energy levels associated with these lines have not, as yet, been identified. However, it is reasonable to expect that many of these are two-electron excitation levels.

Some of the autoionized lines which have been observed in Ar are shown in the bottom spectrum of Fig. 1 for the 420-470Å region. (The first ionization potential for Ar is 15.76 eV.) An Ar pressure of 0.01 mm Hg, and an absorption path length of approximately 105 cm were used. The prominent series of autoionizing lines in this spectrum, due probably to transitions $3s^23p^6\,{}^1S_0$-$3s3p^6np\,{}^1P_1{}^\circ$, is different in character from the prominent series in Ne and He. The Ar series has the appearance of an emission series. These lines must be thought of as discrete decreases in absorption at the line positions, there being no accompanying region of pronounced increase in absorption. Fano[14] has indicated that such shapes are allowable from his theory as a special case of the general interference phenomena when the wave functions and interaction parameters have appropriate values.

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[1]H. Beutler, Z. Physik 93, 177 (1935).
[2]W. R. S. Garton and K. Codling, Proc. Phys. Soc. (London) A75, 87 (1960).
[3]R. W. Ditchburn and R. D. Hudson, Proc. Roy. Soc. (London) A256, 53 (1960).
[4]Po Lee and G. L. Weissler, Proc. Roy. Soc. (London) A220, 71 (1953).
[5]Po Lee and G. L. Weissler, Phys. Rev. 99, 540 (1955).
[6]R. W. Ditchburn, Proc. Phys. Soc. (London) A75, 461 (1960).
[7]N. N. Axelrod and M. P. Givens, Phys. Rev. 115, 97 (1959).
[8]N. Wainfain, W. C. Walker, and G. L. Weissler, Phys. Rev. 99, 542 (1955).
[9]D. J. Baker, Jr., D. E. Bedo, and D. H. Tomboulian, Phys. Rev. 124, 1471 (1961).
[10]R. Whiddington and H. Priestley, Proc. Roy. Soc. (London) A145, 462 (1934); H. Priestley and R. Whiddington, Proc. Leeds Phil. Lit. Soc., Sci. Sect. 3, 81 (1935-1940).
[11]S. Silverman and E. H. Lassettre, Ohio State University Research Foundation Report No. 9 (unpublished); see also reference 14.
[12]Ta-You Wu, Phys. Rev. 66, 291 (1944).
[13]B. H. Bransden and A. Dalgarno, Proc. Phys. Soc. (London) A66, 904 (1953).
[14]U. Fano, Nuovo Cimento 12, 156 (1935); Phys. Rev. 124, 1866 (1961).
[15]J. Cooper, U. Fano, and F. Prats, following Letter [Phys. Rev. Letters 10, 518 (1963)].

CLASSIFICATION OF TWO-ELECTRON EXCITATION LEVELS OF HELIUM

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Madden and Codling[1] describe a series of autoionizing levels of He, in the 60- to 65-eV range, which converges to the $n=2$ level of He^+. The classification of these levels as ${}^1P$ appears obvious. However, since the $n=2$ level of He^+ is degenerate ($2s$ and $2p$), two separate He series might be expected to converge to this limit, namely, $2snp$ and $2pns$. These series have a common lowest level with $n=2$. The probabilities of dipole excitations from the ground state $1s^2$ to $2snp$ or $2pns$ should be comparable for reasons indicated below. Therefore the observation of a single series indicates that the classifications $2snp$ and $2pns$ do not constitute an appropriate zero-order approximation.

Because the states $2snp$ and $2pns$ are nearly degenerate, the electron-electron interaction will, if sufficiently strong, remove their degeneracy in the zeroth step of perturbation treatment and replace the symmetrized independent-electron wave functions $u(2snp)$ and $u(2pns)$ with the pair[2]

$$\psi(2n\pm)=\{u(2snp)\pm u(2pns)\}/\sqrt{2}. \quad (1)$$

Indeed, a calculation with screened hydrogenic wave functions shows that the electron interaction matrix element $(2s3p\,|\,V\,|\,2p3s)\sim 1$ eV, whereas $E_{2s3p}-E_{2p3s}\sim 0.1$ eV.

This Letter points out the following properties

of the new zero-order states[1]:

(a) The + (-) quantum number corresponds to radial motions of the two electrons in (out of) step with one another.

(b) The $2s2p$ state belongs to the + classification.

(c) The + level of each pair lies presumably above the - level.

(d) The optical transition from $1s^2$ to the - level is quasi-forbidden.

(e) The radiationless transition from the + levels to the $1skp$ continuum is quasi-forbidden.

(f) The radiationless transition from the - levels is also quasi-forbidden, by the same mechanism that leads to (d).

According to (d) and (e), the narrow observed levels are classified as +; future observation of weak and very narrow - levels is conceivable.

Consider at the outset that the wave functions $u(2snp)$ and $u(2pns)$ are nearly equal when both electrons lie within the region that the electrons occupy in their ground state $1s^2$. (The two states differ by several eV in the energy allotted to the s and p electrons in an independent-electron approximation, but this energy difference is negligible as compared to the potential energy within the $1s^2$ region.) Therefore the wave function of the - states nearly vanishes in this region, and the dipole moment connecting it to the $1s^2$ state is very small [point (d)].

To characterize qualitatively the + and - states, one may consider an analog of the radial motion of two excited electrons, consisting of two identical pendulums suspended from points at the same level on a vertical wall so that each can perform a periodic motion by rebounding from the wall. Introduction of a weak coupling between the pendulums singles out to two modes of joint periodic oscillation. In one mode (+) the pendulums approach and hit the wall simultaneously, as the He electrons may simultaneously overlap the $1s^2$ orbital in the + state; in the other mode (-) one pendulum hits the wall when the other is farthest from it. If the two pendulums' distances from the wall are r_1 and r_2, $r_1^2 \pm r_2^2$ is a periodic function of time and $r_1^2 \mp r_2^2 =$ const in the $\pm$ mode. In a quantum mechanical treatment of the two pendulums, their joint wave function may be mapped on the (r_1, r_2) plane and will have nodal lines generally perpendicular to the classical trajectory of the representative point (r_1, r_2) which lies along (across) the diagonal $r_1 = r_2$.

A corresponding analysis of the radial motion of the He electrons may be obtained starting from the Breit[3] form of the ^1P wave function $\psi(r_1, r_2)$ $= F(r_1, r_2, \theta_{12})Y_{1m}(\theta_2, \varphi_2) + F(r_2, r_1, \theta_{12})Y_{1m}(\theta_1, \varphi_1)$, expanding $F(r_1, r_2, \theta_{12}) = \sum_k F_k(r_1, r_2)P_k(\cos\theta_{12})$ and mapping $F_0(r_1, r_2)$. For purposes of orientation F_0 has been constructed with products of hydrogenic radial wave functions R_{nl} as follows:

$$F_0(22+; r_1, r_2) = F_0(2s2p; r_1, r_2) = R_{20}(r_1)R_{21}(r_2), \quad (2)$$

$$F_0(2n\pm; r_1, r_2) = R_{ns}(r_1)R_{2p}(r_2) \pm R_{2s}(r_1)R_{np}(r_2). \quad (3)$$

(Note that $F_k = 0$ for $k \neq 0$ in any independent-electron approximation.)

Figure 1 shows the maps for $n = 2, 3$; $n = 4$ has also been studied. As a criterion to classify the the type of motion, one may take the number of nodal lines crossed by the line $r_1 = r_2$ between the origin and ∞. This number appears to be $n - 2 \pm 1$ for $2n\pm$. Another criterion consists of estimating the mean value of $|r_2 - r_1|$, which is small (large) for + (-) states [point (a)]. Thus the maps exhibit some of the characteristics predicted for the two-pendulum model, with allowance for the distortion due to the asymmetry between s and p waves.

The values of transition matrix elements depend primarily on the overlap of the F_0 functions of the initial and final states. Observe particularly the magnitude of F_0 in the range of overlap with the $1s^2$ wave function, namely, $(r_1, r_2) \lesssim 1$: Here the $F_0(2n+)$ are rather large and the $F_0(2n-)$ small, as anticipated [point (d)]. (One may say that, if the two electrons are excited simultaneously out of the $1s^2$ orbital near the nucleus, their subsequent radial oscillations will remain in step.) The similarity of 22+ and 23+ in this respect justifies point (b). The smallness of $F_0(2n-)$ all along the line $r_1 = r_2$, as well as throughout $(r_1, r_2) \lesssim 1$, indicates that the electron repulsion is minimized in this state, and thus leads to point (c).

In first-order perturbation theory the rate of autoionization will depend on the overlap of $F_0(2n\pm)$ with the continuum function $F_0(1skp)$ $= R_{10}(r_1)R_{k1}(r_2)$, whose successive maxima and minima lie along the r_2 axis. The first node of this function lies at $r_2 \sim 2$ for energies of the order of 60 eV and thus coincides with the first maximum of $F_0(2n+)$ and with the first node of $F_0(2n-)$ on the r_2 axis. Coincidence of a node of one state with a maximum of the other causes a cancelation of positive and negative contributions to integrals over space coordinates and thereby reduces the transition matrix elements between $2n+$ and $1skp$ [point (e)]. The coincidence of the first nodes of $2n-$ and $1skp$ tends instead to raise the value of the corresponding matrix element, but this matrix element is never-

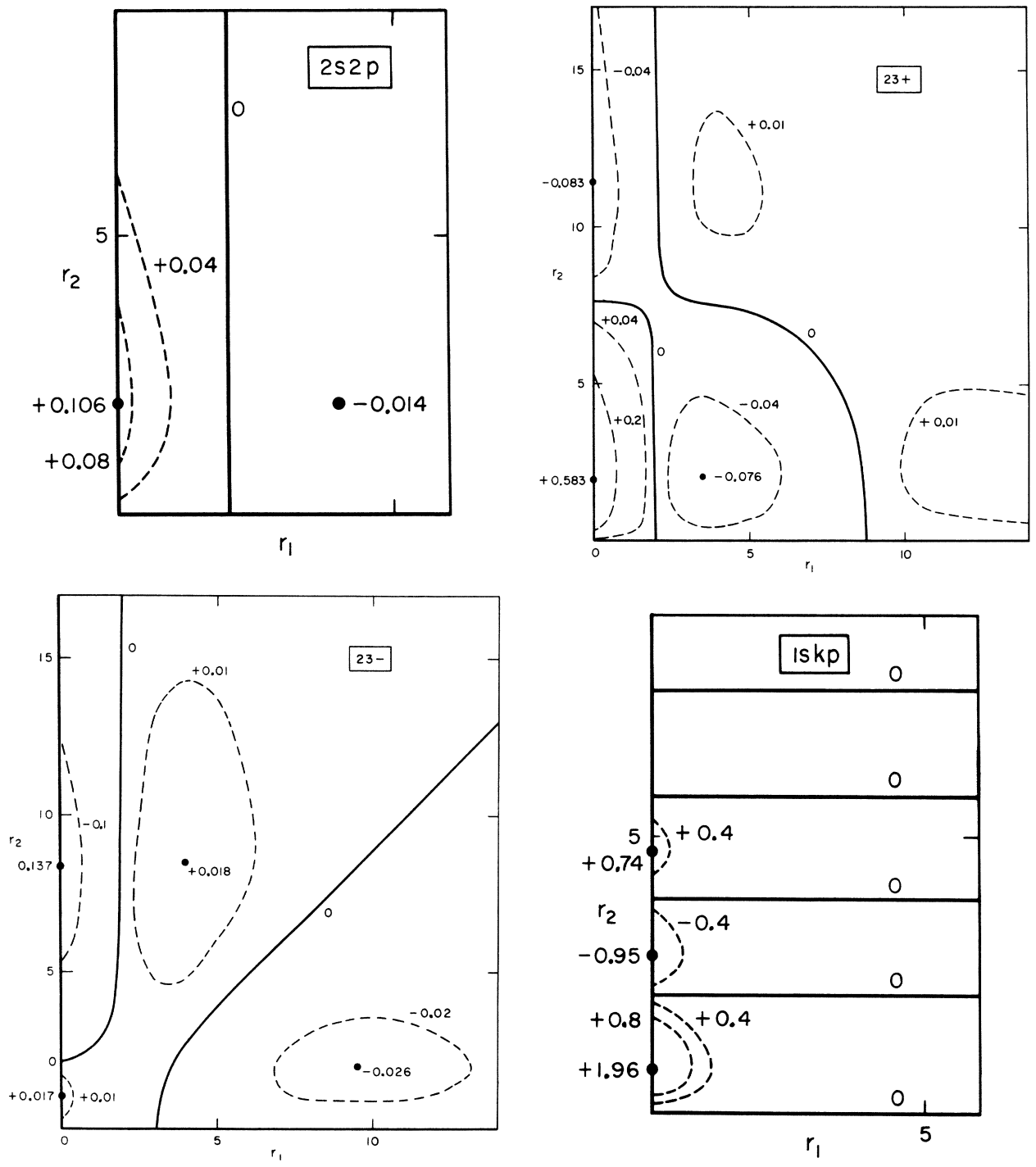

FIG. 1. Contour maps of an approximate $F_0(r_1, r_2)$ for various states. The kinetic energy of the kp electron corresponds to level 61 eV above the $1s^2$ of He.

theless small because of the small value of $2n$- throughout the region of the first maximum and first minimum of $1skp$. No substantial contribution accrues to either matrix element from the range of larger values of r_2, because $1skp$ oscillates too fast in this region. In physical language, an electron can be propelled by Coulomb repulsion to the high speed of the free kp state only in a region sufficiently close to the other electron. Thus autoionization of the - states appears to be

quasi-forbidden by much the same circumstance which makes its excitation unlikely [point (f)].

With regard to the possible relevance of the ± classification to atoms other than He, the complete degeneracy of the series limit is, of course, peculiar to two-electron atoms or ions. However, all states with two highly excited electrons will resemble the corresponding states of He in the same sense as all states of one-electron excitation belonging to Rydberg series resemble the H states. Doubly excited states may arise by mechanisms other than optical excitation. In particular, bombardment of neutral atoms with electrons of energy comparable to the ionization threshold might often result in the capture of the incident electron to form a two-electron state with an excited atomic electron. Evidence for this process is seen in the recent discovery of a resonance in elastic scattering by He at 19.3 eV,[4] in the theoretical prediction of similar resonances for H,[5] and in the behavior of the polarization of electron-induced light at a few volts above threshold, as interpreted by Baranger and Gerjuoy.[6]

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[1]R. Madden and K. Codling, preceding Letter [Phys. Rev. Letters 10, 516 (1963)].

[2]Normalization of the u to be real and positive when both electrons lie near the nucleus makes the ± sign physically meaningful.

[3]G. Breit, Phys. Rev. 35, 569 (1930).

[4]G. J. Schultz, Phys. Rev. Letters 10, 104 (1963); this resonance and a similar one in Ne have also been observed by J. A. Simpson (private communication).

[5]P. G. Burke and H. M. Schey, Phys. Rev. 126, 147 (1962); R. Damburg and R. Peterkop, Proc. Phys. Soc. (London) 80, 1073 (1962); A. Temkin, Phys. Rev. Letters 10, 22, 268 (1963).

[6]E. Baranger and E. Gerjuoy, Proc. Phys. Soc. (London) 72, 326 (1958). For recent experimental results see R. H. McFarland and E. A. Soltysik, Phys. Rev. 127, 2060 (1962), and 128, 1758 (1962); R. H. Hughes, R. B. Kaye, and L. D. Weaver, Phys. Rev. 129, 1630 (1963).

ISOTOPE SHIFT AND SATURATION BEHAVIOR OF THE 1.15-μ TRANSITION OF Ne†

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This Letter gives a summary of the results of an experiment with He-Ne optical maser in which the isotope effect together with the saturation behavior of one of the Doppler-broadened optical transitions of Ne, the $2p_4 - 2s_2$ transition[1] at 1.15 μ, are studied. An accurate value of the isotope shift due to Ne^{20} and Ne^{22} isotopes is measured, and the saturation parameters related to pressure effects and spontaneous radiative decays are determined.

The power output and the exact oscillation frequency of an optical maser depend on the saturation behavior of the atomic transition. These dependences may, hence, be used to study the details of the atomic line shape. This technique was adapted to the study of the 1.15-μ transition of Ne atoms. From the early stages of the experiment, it became evident that the slight distortion of the Doppler line shape due to the presence of 9% Ne^{22} with a small isotope shift introduced a considerable amount of ambiguity in the interpretation of the observed effects. In order to measure the isotope shift and to determine its effect on the over-all saturation of the resonance, two separate He-Ne optical masers were constructed containing isotopically enriched samples of Ne^{20} and Ne^{22}, respectively. The value of the isotope shift was determined as follows. Each maser was allowed to oscillate on a single axial mode. The oscillation frequency of each maser was then adjusted to coincide with the center of its respective Doppler-broadened transition. To achieve this, use was made of the saturation behavior to be described below. The outputs of the two masers were then heterodyned on the photocathode of a photomultiplier as described previously.[2] The difference frequency was measured by analyzing the frequency of the beat note at the output of the photomultiplier using a radio receiver. The value of the isotope shift was thus determined to be 261 ± 3 Mc/sec at 0.1 mm Ne, 1.0 mm He partial pressures, with the heavier isotope lying on the high-frequency side. In view of some unknown pressure shifts which are still subject to further studies, the quoted error is a conservative estimate.

The optical field within the maser resonators used in our experiments has the form of a stand-