Vacuum Ultraviolet Radiation and Jesse Effects in the Noble Gases*

G. S. Hurst, T. E. Stewart, and J. E. Parks
Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
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When charged particles interact with the noble gases, energy is radiated in the vacuum ultraviolet (VUV) region of the electromagnetic spectrum. Some recent measurements of the absolute yield of VUV energy per cm of proton track length show that this quantity $d\epsilon/dx$ is an appreciable part of the total stopping power dE/dx. Here, we show by direct comparisons and by arguments based on the principle of conservation of energy that the Jesse effect (Penning ionization) in helium, neon, and argon is associated with radiation energy in the VUV region.

I. INTRODUCTION

When a swift charged particle interacts with a gas, a macroscopic parameter, the stopping power dE/dx, is frequently used to describe the energy losses involved in the interaction. This macroscopic parameter includes (i) the energy lost by the particle in creating ions, (ii) the energy lost in the excitation of atomic states, (iii) the energy dissipated in the form of subexcitation electrons, and (iv) others. In other words, stopping power can be expressed as

$$\frac{dE}{dx} = n_i I + \sum N_j \epsilon_j + n_i E_s + E_x \ . \tag{1}$$

In Eq. (1), n_i is the number of ion pairs formed per cm of track, I is the ionization potential of the atom, N_j is the number of excited states formed per cm which have energy ϵ_j, and E_s is the average energy of subexcitation electrons. The term $\sum N_j \epsilon_j$ represents the total energy spent in excitation processes and includes both radiation from excited states and, for example, energy converted to ground-state repulsive potentials in two-body systems, arising from Franck-Condon processes. The term E_x is energy dissipation not explicitly observed, but is required to balance Eq. (1).

Research over the past decade suggests that for the interaction of protons with the noble gases, the above list is given roughly in the order of importance. Very recent measurements[1] indicate that for 4-MeV protons interacting with the noble gases, the energy term due to excited states can be an appreciable fraction of the total stopping power and is great enough to be responsible for Jesse effects in the noble gases. To develop our thesis, it is necessary to discuss the nature of excited atomic states and their destruction by atomic collisions.

It is useful to classify the excited atomic states as (a) metastable, (b) resonance, and (c) cascading. By (c) we shall mean a state which cascades with sufficient frequency that anomalously long system lifetimes are not generated by imprisonment, as occurs in case (b). At pressures of interest (greater than 100 Torr) we can disregard the diffusion of excited states to the walls of a container even in the case of long-lived metastable states or trapped resonance photons. Therefore, we properly assume that the fate of the excited species in the cases (a) and (b) will depend on collision processes. In the case of (c), particularly at the lower pressures, it is conceivable that an atom will radiate before collision occurs.

An atom excited to a metastable state may collide with a ground-state atom to be converted to a radiating state or converted to another metastable state. Alternatively, collision of metastable states with ground-state atoms may stimulate photon emission. Metastable states are often destroyed in three-body processes[2,3] which presumably form a diatomic molecule in a vibrational eigenstate. This molecule in turn could emit electromagnetic radiation[4] in the form of a continuum or it could be relaxed by additional collisions with atoms.

Atoms excited to a resonance state emit photons which are imprisoned[5] sufficiently that one may consider it of long lifetime. In this respect, a resonance state has most of the features of a metastable state. Thus an atom in a resonance state, in effect, may collide with a ground-state atom to be converted to a metastable state. More likely, the two-body collision produces a Franck-Condon effect[6] in which the ground repulsive state of the two-body system is at a position of positive potential energy. Thus, the photon emitted is nonresonant and escapes from the system. Three-body collisions could also proceed as above to produce molecular emission through a molecular intermediate.

It is clear that, regardless of the detailed sequence of events, excited states created by the interaction of charged particles with atoms can emit continuous spectra of photons through various collision mechanisms. Moreover, in the case of noble gases, these radiations will be in the vacuum ultraviolet (VUV) region of the spectrum. A recent paper[1] reported the spectra produced by 4-MeV pro-

ton interactions with all of the stable noble gases and showed measurements of the energy radiated per cm of proton pathlength. This radiant energy per cm of track length, $d\epsilon/dx$, was an appreciable part of the stopping power dE/dx.

We wish to summarize this new information in context with total ionization experiments in which W values (electron volts per ion pair) are obtained. We shall be particularly interested in commenting on the connection between the VUV spectroscopic measurements and the so-called Jesse effect in the noble gases.

II. METHOD

We have previously described[6] a method for investigating the interaction of protons with gases. In this method, protons lose a known amount of energy per cm in a gas (ranging in pressure from about 10 to 1500 Torr) and the light emitted in the VUV region of the electromagnetic spectrum is measured with a $\frac{1}{2}$-m scanning monochromator. More recently[1] we have shown that the amount of energy radiated from a given length of track can be determined absolutely. This determination involves using a calibrated channel electron multiplier in a simple geometry experiment. Also required are the transmission of a LiF window and the reflectivity of the diffraction grating. References 1 and 6 may be consulted for further experimental details.

Specifically, what we have measured with the above methods is the quantity

$$\frac{d\epsilon}{dx}=\int \epsilon(\lambda)\Phi(P,\lambda)\,d\lambda \ , \qquad (2)$$

where $\Phi(P,\lambda)\,d\lambda$ is the total number of photons emitted per cm of track in the region between λ and $\lambda+d\lambda$ due to one proton passing through the gas at pressure P, and $\epsilon(\lambda)$ is the energy of the photon of wave-length λ. According to the discussion in Sec. I, the measured quantity $d\epsilon/dx$ is not identical with the second term $\sum N_j\epsilon_j$ in Eq. (1) but is closely related to it.

Under the conditions of our experiments, we are able to measure the intensity as a function of pressure $I(P)$. The quantity $I(P)/P$ is essentially proportional to the energy radiated per unit of proton energy loss, since the stopping power is constant. Therefore, one may reason that the function $I(P)/P$ will be well-behaved. Suppose we are looking at continuous emission arising from collisions of excited atoms with ground-state atoms. At low pressures, excited atoms or resonance radiation will escape to the walls of the apparatus. As the pressure increases, $I(P)/P$ will approach an asymptotic limit or it will reach a maximum and decline in a way that depends on collision processes in competition with radiation. For example, for two-body Franck-Condon processes leading to an observed continuum, $I(P)/P$ is asymptotic. If three-body collisions compete with two-body effects in a nonradiative pathway, $I(P)/P$ will reach a maximum. The collision considerations above lead us to the conclusion that the measured $d\epsilon/dx$ at a pressure where $I(P)/P$ is asymptotic or a maximum will be a lower limit to the term $\sum N_j\epsilon_j$. Furthermore, $d\epsilon/dx$ is a lower limit because the potential energies in repulsive states connected with radiation processes are not included in $d\epsilon/dx$.

TABLE I. Measured values of the radiant energy per proton pathlength, $d\epsilon/dx$, compared with dE/dx, the stopping power.

Gas	Pressure (Torr)	$\frac{d\epsilon}{dx}$ keV/cm	$\frac{dE}{dx}$ keV/cm	$\frac{d\epsilon/dx}{dE/dx}$
Helium	400	2.2	9.1	0.24
Neon	200	4.7	17.2	0.27
Argon	400	16.0	55.4	0.29
Krypton	400	10.5	84.4	0.12
Xenon	300	8.1	87.1	0.09

TABLE II. Basic energy values (eV) appropriate to fast low-mass charged-particle ionization and excitation of the stable noble gases.

Gas	W	W_m	I	$\frac{I}{W}$	$(1-I/W)$	ϵ_{jo}	$\frac{E_J}{dE/dx}$
He	46.0[a]	30.0[b,c]	24.5	0.53	0.47	19.8	0.24
Ne	39.3[c]	25.2[d]	21.5	0.55	0.45	16.6	0.24
Ar	26.4[c]	20.5[e,c]	15.7	0.59	0.41	11.6	0.13
Kr	24.0[f]	22.3[f]	13.9	0.58	0.42	9.9	0.04
Xe	21.5[g]	...	12.1	0.56	0.44	8.3	...

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[b]See Ref. 7.

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III. RESULTS AND DISCUSSION

Measured[1] values of the quantity $d\epsilon/dx$ for all of the stable noble gases are reproduced in Table I, where a comparison is made with the total stopping power dE/dx. The column $d\epsilon/dx$ indicates that a rather large fraction of the total energy lost by the protons is radiated out of the gas.

We now wish to discuss the quantity $d\epsilon/dx$ in context with W values and Jesse effects in the stable noble gases. The W value of a gas is defined as the energy lost by a particle divided by the number of ion pairs formed during the energy-loss process.

The usual method for measuring W is to fill the apparatus with gas at sufficient pressure so that the charged particle is completely absorbed. One collects the ionization charge created by a known number of particles at known energy. It is now well known that measured W values in noble gases are quite sensitive to impurities. For example, the number of ion pairs increases when a few parts per million of almost any gas is added to helium[7,8]; this increase is referred to as the Jesse effect or as Penning ionization. Jesse effects have also been observed in neon, argon, and krypton; see Table II.

Associated with the Jesse effect is a decrease in the W value of a noble gas. Specifically, if one introduces various impurities into a purified noble gas, it is observed that a minimum W value is found at a certain pressure for each of the impurities. The absolute minimum W_m for the impurity having the largest effect and the W value for the purified noble gases are shown in Table II. Also shown in Table II is the ionization potential of each gas, I, and the energy of the lowest excited state in each noble gas, ϵ_{j0}.

Referring to Eq. (1), let $E_{ex}=\sum N_j\epsilon_j$ and divide by dE/dx; then

$$1=\frac{I}{W}+\frac{E_{ex}}{dE/dx}+\frac{E_s}{W}+\frac{E_x}{dE/dx} \quad . \tag{3}$$

An upper limit on the fraction $E_{ex}(dE/dx)^{-1}$ is therefore

$$\frac{E_{ex}}{dE/dx}\leq 1-\frac{I}{W} \quad , \tag{4}$$

where we have neglected the energy dissipated by subexcitation electrons as well as the last term in Eq. (1). The fraction in the final column of Table I cannot exceed the term $E_{ex}(dE/dx)^{-1}$. Therefore $(d\epsilon/dx)(dE/dx)^{-1}$ will be less than $(1-I/W)$; see Table II.

Another significant comparison is obtained by calculating the fraction of energy lost by the charged particle which must be associated with observed Jesse effects. For this we note that the increase in ion pairs per cm of track due to the Jesse effect is

$$\Delta n_i = dE/dx\left(\frac{1}{W_m}-\frac{1}{W}\right) \quad . \tag{5}$$

Thus, $E_J\ (dE/dx)^{-1}$, the fraction of the total energy lost available for the Jesse effect, is approximately

$$E_j/\frac{dE}{dx}=\epsilon_{j0}\frac{W-W_m}{WW_m} \quad , \tag{6}$$

in which ϵ_{j0} is the energy of the lowest-lying excited state of the noble-gas atoms. The assumptions leading to Eq. (6) imply that the Jesse effect is produced as a consequence of energy transfer from excited atomic states to an atom or molecule which is then ionized. We have not specified whether the atomic states are resonance or metastable; however, in using ϵ_{j0} as defined, we imply that the energy difference is not critical to this assignment. We note, in support, that for all of the noble gases there is a group of lowest excited states, two of which are resonance and two of which are metastable, and they are all comparable in energy.

TABLE III. Comparison of various energy terms involved in the interaction of 4-MeV protons with the stable noble gases.

Gas	$\frac{d\epsilon/dx}{dE/dx}$	$\frac{E_J}{dE/dx}$	I/W	Sum
Helium	0.24	0.24	0.53	1.01
Neon	0.27	0.24	0.58	1.09
Argon	0.29	0.13	0.59	1.01
Krypton	0.12	0.04	0.58	0.74
Xenon	0.09	...	0.56	...

In the last column of Table II, the fraction $E_J(dE/dx)^{-1}$ is shown. This fraction can be used in a number of interesting comparisons, summarized in Table III. First, we see from the second and third columns of Table III that, for each gas, enough energy is found to be associated with radiation from excited states to account for the Jesse effects. In fact, column 2 is less than the energy residing in the long-lived excited states because potential energy associated with ground repulsive states in continua formation has not been included. We now look at energy bookkeeping by means of Eq. (3). By neglecting completely the last two terms in Eq. (3), the sum of the terms of Table III is approximately unity for He, Ne, and Ar. We know that important energy sources have been neglected (potential energy of ground repulsive states, subexcitation electrons, and radiationless transitions). For example in the case of argon, these three terms add at least 0.10 to the sum in Table III. Therefore, it is not reasonable to add the energies associated with Jesse effects as if they were independent of the term $d\epsilon/dx$. In other words, columns 2 and 3 in Table III are not independent displays of excited states. We conclude that the same atomic states which lead to continua formation in fact lead to Jesse effects when impurities are present.

From direct comparison and from conservation of energy we have argued that Jesse effects in helium, neon, and argon are associated with radiation energy which can be measured in the VUV region. By inference, we speculate that the statement holds for the noble gases generally. This argument has not resolved the question of the nature of the excited states. Metastable states are candidates along with resonance states. However, with argon it has been argued[6] that the observed VUV continua originate from atomic resonance states, and thus in this case Jesse effects originate there also.

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Broadening of Highly Excited Atomic Levels in an Atmosphere of Alkali-Metal Atoms

L. P. Presnyakov*
Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colorado 80302
(Received 13 April 1970)

Interactions between a highly excited atom and another atom in the ground state with nonzero electron affinity lead to a splitting of highly excited levels. This situation provides a mechanism which modifies spectral lines of a Rydberg series. Electronic transitions between components of the quasi molecular level due to interatomic motion are analyzed on the basis of the adiabatic theory. The width of a spectral line near the edge of the series is expressed in terms of the electron affinity of the perturbing atom and other calculable quantities. Comparison of the present theoretical result and experimental data for K and Cs shows that this mechanism is as significant for linewidths as the Fermi process for line shifts.

I. INTRODUCTION

The theory of the pressure shift of the absorption spectral lines near the edge of a series was originated by Fermi.[1] According to Fermi, the shift of the absorption line corresponding to the transition from the ground-state level to a level with a large principal quantum number n is due to two effects: elastic scattering of the atomic electron in the state n by perturbing particles and polarization of the perturbing particles situated inside the atomic volume $(a_0 n^2)^3$, where a_0 is the atomic unit of length. These effects make additive contributions, and at relatively low pressure the polarization effect is very small. The original Fermi formula for the line shift Δ_{sc} (in $\sec^{-1}$) due to the scattering effect has the form

$$\Delta_{sc} = \pm (\hbar/m)[\pi\sigma(0)]^{1/2} N \ , \qquad (1)$$

where m is electron mass, $\sigma(0)$ is the effective cross section for elastic scattering of an orbital electron (which is considered as a quasi-free-electron) by a perturbing atom in the limit of zero velocity, and N is the concentration of the perturbing atoms. Firsov[2] showed that Eq. (1) is valid only if the most probable value of the electron velocity in the state n is so small that the main contribution to the elastic cross section is made by the s-wave scattering only.

The Fermi theory was completed by Alekseev and Sobelman.[3] They showed that Fermi's idea of elastic scattering of a quasi-free orbital electron corresponds to the well-known impulse approximation, and they obtained expressions for both the shift Δ_{sc} and the width γ_{sc} of spectral lines:

$$\Delta_{sc} = N \frac{\hbar}{m} \int_0^\infty \left(\frac{\pi}{q} \sum_l (2l+1) \sin 2\delta_l \right) w(q)\, dq \ , \qquad (2)$$

$$\gamma_{sc} = N \frac{\hbar}{m} \int_0^\infty \left(\frac{4\pi}{q} \sum_l (2l+1) \sin^2 \delta_l \right) w(q)\, dq \ . \qquad (3)$$

Here $\delta_l(q)$ is the phase of the lth partial wave for elastic scattering of the electron of momentum q by the perturbing atom, and $w(q) = q^2 |G_{nl}(q)|^2$, where $G_{nl}(q)$ is the wave function of an orbital electron in the momentum representation. Equation (2) reduces to Fermi's result (1) in the limiting case of the s-wave scattering only. Alekseev and Sobelman[3] point out that exchange interaction should be taken into account as well.

Until recently, experimental results on broadening and shift of the absorption spectral lines near the edge of the series concerned alkali-metal atoms in an atmosphere of inert gases only. In this case, the theoretical calculations of Alekseev and Sobelman are in very good agreement with experimental results.[3-5] The widths and shifts do not depend on the principal quantum number n for n