

nucleus by observing the angular correlation of the emitted particles as a function of time.<sup>3</sup> One would expect a correlation to persist to times of the order of the total spin relaxation time  $T_2$  of the intermediate nuclei  $j$  in the sample, perhaps milliseconds or better.<sup>3</sup> We show here how the theoretical delayed coincidence correlation function describing the effect of the magnetic field is to be obtained from the usual correlation function.

The equations of the Weisskopf-Wigner treatment of the decay<sup>4</sup> can be made to yield a function  $W(\Omega_1; \Omega_2; t-t')$  which can be interpreted as the rate (steradian<sup>-2</sup> sec<sup>-1</sup>) at which particles of the second transition are emitted at time  $t$  with direction  $\Omega_2 \equiv \theta_2, \varphi_2$  when the preceding particle in the cascade was emitted at time  $t'$  with direction  $\Omega_1 \equiv \theta_1, \varphi_1$ , the initial nuclei  $j_1$  being randomly oriented at time zero. As indicated, this rate comes out to be a function of  $t-t'$  only, and we refer to it as the delayed coincidence angular correlation function.<sup>5</sup> If  $\omega_0$  is the circular Larmor frequency of the intermediate nucleus  $j$  in the external magnetic field (along the  $z$ -axis), then the delayed coincidence correlation is given by

$$W(\theta_1, \varphi_1; \theta_2, \varphi_2; t) = 4\pi\tau_1 \exp(-t/\tau) \times \sum_{mm'} X_{mm'}(\theta_1, \varphi_1; \theta_2, \varphi_2) \exp(i\omega_0(m-m')t), \quad (1)$$

where

$$X_{mm'}(\theta_1, \varphi_1; \theta_2, \varphi_2) = (2j_1+1)^{-1} (2\pi/\hbar)^2 \rho_1 \rho_2 \mathcal{S} \sum_{m_1 m_2} \times [(j_2 m_2 | \Omega_2, \sigma_2 | j m) (j m | \Omega_1, \sigma_1 | j_1 m_1) \times (j_2 m_2 | \Omega_2, \sigma_2 | j m')^* (j m' | \Omega_1, \sigma_1 | j_1 m_1)^*]. \quad (2)$$

In Eq. (1),  $\tau_1$  and  $\tau$  are the lifetimes of the initial and intermediate nuclei, respectively, and in Eq. (2),  $\mathcal{S}$  is the invariant summation over the spin coordinates  $\sigma_1$  and  $\sigma_2$  of the emitted particles, and  $\rho_1$  and  $\rho_2$  are densities-in-energy of the states of the emitted particles. The usual instantaneous angular correlation<sup>6</sup> is

$$W(\theta_1, \varphi_1; \theta_2, \varphi_2) = \tau W(\theta_1, \varphi_1; \theta_2, \varphi_2; 0) = 4\pi\tau_1 \sum_{mm'} X_{mm'}(\theta_1, \varphi_1; \theta_2, \varphi_2), \quad (3)$$

normalized to  $\int W(\theta_1, \varphi_1; \theta_2, \varphi_2) (d\Omega_1/4\pi) d\Omega_2 = 1$ . It is actually a function only of the angle  $\theta$  between  $\Omega_1$  and  $\Omega_2$ , of course.

The transformation properties of the nuclear matrix elements<sup>7</sup> can be used to obtain

$$X_{mm'}(\theta_1, \varphi_1; \theta_2, \varphi_2) = X_{mm'}(\theta_1, \varphi_1; \theta_2, \varphi_2 + \alpha) \exp(i\alpha(m' - m))$$

for rotations of  $\Omega_2$  by angle  $\alpha$  around the  $z$ -axis. If this is substituted into Eq. (1) with  $\alpha = \omega_0 t$ , one discovers that

$$W(\theta_1, \varphi_1; \theta_2, \varphi_2; t) = (1/\tau) \exp(-t/\tau) W(\theta_1, \varphi_1; \theta_2, \varphi_2 + \omega_0 t). \quad (4)$$

The delayed coincidence angular correlation pattern is just the instantaneous pattern precessing with the intermediate nucleus around the external field.<sup>8</sup> If the instantaneous correlation is given as a power series in  $\cos\theta$ , the corresponding delayed coincidence correlation is obtained as a Fourier series in  $\omega_0 t$  by transforming the instantaneous correlation to a Legendre series.<sup>9</sup>  $W(\theta) = \sum \gamma_l P_l(\cos\theta)$ , and then applying the spherical harmonic addition theorem<sup>9</sup> to

$$P_l(\cos\theta) = P_l(\cos\theta_1 \cos\theta_2 + \sin\theta_1 \sin\theta_2 \cos(\varphi_1 - \varphi_2)).$$

For example, the instantaneous correlation  $W(\theta) = 1 + r \cos^2\theta = 1 + (r/3) + (2r/3)P_2(\cos\theta)$  gives the delayed coincidence correlation (omitting the decay and normalization factors):

$$W(\theta_1, \varphi_1; \theta_2, \varphi_2; t) = 1 + (r/3) + (r/6)(3 \cos^2\theta_1 - 1)(3 \cos^2\theta_2 - 1) + (r/2) \sin 2\theta_1 \sin 2\theta_2 \cos(\varphi_1 - \varphi_2 - \omega_0 t) + (r/2) \sin^2\theta_1 \sin^2\theta_2 \cos 2(\varphi_1 - \varphi_2 - \omega_0 t). \quad (5)$$

In general, the odd harmonics of  $\omega_0 t$  drop out if either counter is in the equatorial plane,  $\theta_1$  or  $\theta_2 = 90^\circ$ ; and there are no oscillating terms at all if either counter is on the axis,  $\theta_1$  or  $\theta_2 = 0^\circ$  or  $180^\circ$ . The relative amplitudes of the various harmonics are complicated functions of  $\theta_1$  and  $\theta_2$ , in general, and the best position for the counters has to be worked out for each case separately. For the correlation (5), one maximum of the first harmonic amplitude relative to the non-oscillating terms occurs when both counters are on the cone:

$$\cos^2\theta_1 = \cos^2\theta_2 = (1/2) - (r/(8+6r)) + O(r^2).$$

It is to be noted that both the instantaneous correlation and the gyromagnetic ratio of nucleus  $j$  can be determined by measuring the distribution of pulses in time in one counter; e.g., if the counter is in the equatorial plane,  $W(90^\circ, \varphi; 90^\circ, \varphi; t) = (1/\tau) \exp(-t/\tau) W(\omega_0 t)$ , where  $W(\theta)$  is the instantaneous correlation.

If the highest power of  $\cos\theta$  in the instantaneous correlation is  $\cos^L\theta$ , the delayed correlation contains harmonics up to  $\cos(L\omega_0 t)$ , and the total gate width  $\Delta t$  should be narrow enough so that these harmonics are not washed out by averaging over the gates: if  $L\omega_0 \Delta t \geq 1$ , gate width correction factors must be introduced. For the same reason, the variation  $(T_2^*)^{-1}$  of the Larmor frequency due to inhomogeneities in the applied magnetic field should be kept small compared to  $(L\tau)^{-1}$ .

I wish to thank Professor A. T. Nordsieck for a discussion of the Weisskopf-Wigner method and Dr. C. P. Slichter for discussions on the nature of the fields in material media.

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<sup>2</sup> Sunyar, Alburger, Friedlander, Goldhaber, and Scharff-Goldhaber, Phys. Rev. **79**, 181 (1950).

<sup>3</sup> Bloembergen, Purcell, and Pound, Phys. Rev. **73**, 679 (1948).

<sup>4</sup> V. Weisskopf and E. Wigner, Z. Physik **63**, 54 (1930).

<sup>5</sup> G. Goertzel deals essentially with  $\int_0^\infty W(\Omega_1; \Omega_2; t) dt$  in his investigation of the effect of hyperfine structure interactions in  $\gamma$ - $\gamma$  angular correlations; Phys. Rev. **70**, 897 (1946). We assume that there are no such fields at the nucleus in the liquid sample.

<sup>6</sup> D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **79**, 323 (1950).

<sup>7</sup> S. P. Lloyd, Phys. Rev. **80**, 118 (1950).

<sup>8</sup> This was suggested by Professor M. Goldhaber (private communication).

<sup>9</sup> The necessary formulas can be found in W. E. Byerly, *Fourier Series and Spherical Harmonics* (Ginn and Company, 1893), pp. 179, 211, 199.

## Electron Density and Light Intensity Decay in Cesium Afterglows\*

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THE decay of the electrodeless discharge through purified cesium has been studied by the previously described techniques<sup>1</sup> of microwave electron density, optical spectrographic, and optical light intensity measurements as a function of time after termination of the exciting fields. In the pressure range 0.02 to 0.1 mm, we found that the electron density,  $n$ , decays exponentially with a time constant,  $\tau_1$ , increasing with pressure from 0.4 millisecond at 0.02 mm to about 1 millisecond at 0.1 mm. A minimum in the light intensity is found at 40 microseconds after cut-off, followed at about 80 microseconds by a broad maximum. The ratio  $I_{\max}/I_{\min}$  is about 8 at the lowest pressure and decreases with pressure. Higher input power during the discharge moves the maximum to shorter times. After the maximum, the light intensity decay is exponential with an associated time constant  $\tau_2$  consistently closely equal to  $\tau_1/2$ , which shows that the light intensity is proportional to  $n^2$ . Spectrograms show an intense line spectrum during the discharge, the usual CsI spectrum coexisting with a great number of other lines belonging to the excited cesium ion. In the afterglow, however, the spectrum is practically a pure band spectrum, which was not analyzed in detail, but which shows up in the same wavelength range as the bands found previously in absorption in cesium vapor.<sup>2</sup>

The low pressure electron density data follows the behavior expected for ambipolar diffusion quite well. In order to compare the ion mobility data for cesium ions through cesium vapor with previous measurements<sup>3</sup> of the mobility of cesium ions through rare gases, the electron temperature at moderate times in the afterglow at a pressure of 0.036 mm was estimated by a standard kinetic theory calculation as 1400°K. On this basis, our data gives a value of the mobility,  $k_0$ , of about 50 cm<sup>2</sup>/volt-sec calculated for 1-mm pressure and 0°C. This value is rather surprising, since the measured values of the mobility of cesium ions through rare

gases are on the order of  $10^8$ . The formation of molecular ions and the possibility of some mechanism such as charge exchange may be used to explain the low observed value of the effective mobility of cesium ions in cesium.

Above about 0.3-mm pressure, electron density decay follows the recombination law,  $dn/dt = -\alpha n^2$ , very well. The recombination coefficient,  $\alpha$ , has a value of the order of  $10^{-7}$  cm<sup>3</sup>/ion-sec. At constant temperature this coefficient is found to be pressure dependent. At 270°C, for instance,  $\alpha$  varies from  $3.5 \times 10^{-7}$  at 0.32-mm pressure to  $1.2 \times 10^{-6}$  at 0.68 mm. At 285°C  $\alpha$  varies from  $3.5 \times 10^{-7}$  at 1 mm to  $1.45 \times 10^{-6}$  at 2.6 mm. At times in the afterglow greater than 400 microseconds the light intensity is again found to be proportional to the square of electron density. At shorter times, however, this relationship does not hold, indicating that recombination is initially somewhat slower. The use of simple optical filters shows that the red portion of the spectrum decays initially somewhat faster than the violet. Spectrograms show much less of the cesium ion line spectrum during the discharge than was found at low pressures. The afterglow spectrum is again almost purely molecular.

Our data are in agreement with the following mechanism for recombination-type electron removal in the afterglow. Cesium molecular ions are formed in triple collisions between an atomic ion and two neutral molecules. This is followed by recombination between molecular ions and electrons. This process results in the formation of excited molecules which radiate a band spectrum in the afterglow, thus making dissociative recombination an unlikely explanation for the high values of recombination coefficient observed. The initial maximum of the light intensity as observed at low pressure would then correspond to the initial increase of molecular ion concentration (since the concentration of such ions during the discharge is probably small, as evidenced by the line spectrum observed) followed by a decrease due to diffusion to the walls and recombination. It is quite possible that some equilibrium ratio between the density of atomic and molecular ions develops in the afterglow. This ratio would depend on pressure, a larger proportion of the ions being molecular at higher pressures. This would explain the observed pressure variation of  $\alpha$  if we assume also that the recombination coefficient for molecular ions is the limiting value observed at high pressure and that the recombination coefficient for atomic ions is much smaller.

No dependable values for the absolute light intensity were obtained in this work (as have been obtained in this laboratory for several other gases), because the spectrograms showed that most of the energy was concentrated in the infrared, beyond the range of our photomultiplier equipment.

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## Charge Renormalization in the Hartree Approximation\*

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THE field equations and canonical commutation relations of the Dirac-Maxwell fields are

$$\{\gamma_{\mu}[(\partial/\partial x_{\mu}) - ie_0 A_{\mu}^0/\hbar] + \kappa\}\psi(x) = 0, \quad (1)$$

$$\square A_{\mu}^0 = J_{\mu}^0 \text{ext}(x) + \frac{1}{2}ie_0[\psi^{\dagger}(x), \gamma_{\mu}\psi(x)], \quad (2)$$

$$[\psi^{\dagger}(\mathbf{r}, t), \psi(\mathbf{r}', t)]_{\pm} = \gamma^{\delta}\delta(\mathbf{r} - \mathbf{r}'), \quad (3)$$

$$[A_{\mu}^0(\mathbf{r}, t), \partial A_{\mu}^0(\mathbf{r}', t)/\partial t] = i\hbar\delta(\mathbf{r} - \mathbf{r}'), \quad (4)$$

in which  $J_{\mu}^0 \text{ext}(x)$  is the external charge and current distribution. If one uses the Hartree approximation of replacing the electron-positron current operator  $J_{\mu}^0(x) = \frac{1}{2}ie_0[\psi^{\dagger}(x), \gamma_{\mu}\psi(x)]$  by its

expectation value  $\langle J_{\mu}^0(x) \rangle$  in Eq. (2), one obtains

$$\square A_{\mu}^0 = J_{\mu}^0 \text{ext}(x) + \langle J_{\mu}^0(x) \rangle. \quad (5)$$

Any covariant gauge invariant evaluation of  $\langle J_{\mu}^0(x) \rangle$  for the "vacuum" state<sup>1</sup> gives the following form:

$$\langle J_{\mu}^0(x) \rangle_{\text{vac}} = -a \square A_{\mu}^0(x) - \int K^0(x-x') \square' A_{\mu}^0(x') dx' \quad (6)$$

for the parts of  $\langle J_{\mu}^0(x) \rangle$  which are linear in the field strength, and we will neglect all nonlinear effects. In Eq. (6)  $K^0(x-x')$  is an invariant function of  $(x-x')$  which describes the Uehling effect and which is not as singular as a  $\delta$ -function. Combining Eq. (5) and Eq. (6), one obtains

$$(1+a) \square A_{\mu}^0 + \int K^0(x-x') \square A_{\mu}^0(x') dx' = J_{\mu}^0 \text{ext}(x). \quad (7)$$

Carrying out a charge and field renormalization, one obtains

$$\begin{aligned} A_{\mu} &= (1+a)^{-1/2} A_{\mu}^0, & e &= (1+a)^{-1/2} e_0, \\ J_{\mu}^{\text{ext}}(x) &= (1+a)^{-1/2} J_{\mu}^0 \text{ext}, & K(x-x') &= (1+a)^{-1} K^0(x-x'), \\ J_{\mu}(x) &= (1+a)^{-1/2} J_{\mu}^0(x). \end{aligned} \quad (8)$$

Under this renormalization, Eqs. (1) and (5) remain unaltered in form apart from the Uehling terms, but  $e_0$  is replaced by  $e$ , and  $e_0 A_{\mu}^0$  is replaced by  $e A_{\mu}$ . Equation (3) remains unaltered, while (4) becomes

$$[A_{\mu}(\mathbf{r}, t), \partial A_{\mu}(\mathbf{r}', t)/\partial t] = i\hbar(1+a)\delta(\mathbf{r} - \mathbf{r}'). \quad (9)$$

The fact that these equations maintain the same form shows that a charge renormalization will not change the energy levels of atomic systems except for the Uehling terms implied by  $K(x-x')$ , if the experimental value of  $e^2/\hbar$  is used. However, by Eq. (9) we see that the value of Planck's constant for the electromagnetic field will be larger than the Planck's constant of the matter field by the factor  $1+a = (e_0/e)^2$ . We thus see that charge renormalization is not consistent with the canonical formalism, at least in the Hartree approximation. We note, however, that if one insists on evaluating  $a$  so that it is nonvanishing, the error introduced by this can be corrected by reducing the electromagnetic field commutators by the factor  $1/(1+a)$ .

If the commutator relation Eq. (9) is to remain consistent in time with the equations of motion, Eq. (7), one may infer that  $\square A_{\mu}(x) = J_{\mu}^*(x)$ , in which  $J_{\mu}^*(x)$  is the experimental external current distribution. Thus, one obtains

$$J_{\mu}^*(x) + \int K(x-x') J_{\mu}^*(x') dx' = J_{\mu}^{\text{ext}}(x). \quad (10)$$

We now note that the conventional evaluation<sup>2</sup> of  $K(x-x')$  is such that  $J_{\mu}^*(x)$  is not uniquely determined by the fictitious external charge and current distribution  $J_{\mu}^{\text{ext}}(x)$ , because Eq. (10) has nonvanishing solutions when  $J_{\mu}^{\text{ext}}(x)$  is put equal to zero. Since it is  $J_{\mu}^*(x)$  which determines the energy levels of atomic systems, it is convenient to arrange the calculation of the ambiguous quantity  $\langle J_{\mu}(x) \rangle_{\text{vac}}$  so that its parts, which are linear in the field strengths, vanish identically. This can be done.<sup>3</sup>

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<sup>2</sup> J. Schwinger, *Phys. Rev.* **75**, 651 (1949).

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## Diffuse X-Ray Scattering by Disordered Binary Alloys

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THE x-ray scattering from binary alloys of the substitutional type shows two effects due to the degree of order (or disorder): (1) Sharp superlattice reflections when long range order is present; and (2) diffuse scattering which depends on the degree of short range order. Theoretical intensity formulas for these two types of scattering have been given by MacGillavry and Strijk<sup>1</sup>