

Optically Modulated X-Ray Diffraction

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The spatial variation in the response of the electronic charge distribution of an atom to an applied electric field is shown to be observable by means of a measurement of the spontaneous parametric decay of an x-ray pump into the optical region.

The spatial dependence of the change in the electronic charge density of an atom, $\delta\rho(\vec{r})$, induced by an applied optical field is shown to be measurable by a new, nonlinear, x-ray diffraction technique. This previously unmeasurable quantity is intimately related to the linear optical polarizability, the nonlinear optical polarizability, and many other important properties; we anticipate that the knowledge of $\delta\rho(\vec{r})$ will prove to be of significance in atomic and solid state physics.

Consider Thomson scattering of x rays by an atom which is illuminated with an optical field. In the presence of this field, for which we take the form $\vec{E}_0(t) = \frac{1}{2}\vec{E}_0[e^{i\omega_0 t} + \text{c.c.}]$, the electronic charge density of the atom, $\rho(\vec{r}, t)$, may be written in first order as

$$\rho(\vec{r}, t) = \rho_u(\vec{r}) + \frac{1}{2}\delta\rho(\vec{r}, \vec{E}_0)[e^{i\omega_0 t} + \text{c.c.}], \quad (1)$$

where $\rho_u(\vec{r})$ is the unperturbed charge density. If we now scatter x rays of frequency ω_x off this atom, and measure the intensity of the Doppler-shifted upper sideband, we find by inspection that this may be considered as arising from a polarization¹ ${}_{\perp}\vec{\mathcal{P}}(\omega_x = \omega_0 + \omega_x)$, of the form

$${}_{\perp}\vec{\mathcal{P}}(\omega_x = \omega_0 + \omega_x) = (-e/m\omega_x^2) {}_{\perp}\vec{E}_x \text{Re}[e^{i\omega_x t} \int \frac{1}{2}\delta\rho(\vec{r}, \vec{E}_0)e^{-i\vec{Q}\cdot\vec{r}} d\vec{r}]. \quad (2)$$

Here the incoming x-ray field is taken to be $\vec{E}_x(\vec{r}, t) = \frac{1}{2}\vec{E}_x\{\exp[i(\vec{k}_x\cdot\vec{r} - \omega_x t)] + \text{c.c.}\}$, \vec{Q} is the wave vector change upon scattering, and the left subscript \perp implies the projection of $\vec{\mathcal{P}}$ and other vectors normal to the \vec{k} vector of the scattered x-ray field. As we show shortly, Eq. (2) is an essentially exact quantum-mechanical result for the dominant contribution to the nonlinear, sum-frequency polarization induced in an atom in the presence of both an x-ray and an optical field. We also describe an experiment which, with readily available equipment, permits a measurement of this nonlinear polarization as a function of \vec{Q} and, hence, by inversion of Eq. (2), a direct measurement of $\delta\rho(\vec{r}, \vec{E}_0)$.

Direct observation of sum-frequency mixing of a laser and a source of x rays is unattractive because of the low spectral brightness of available spontaneous-emission x-ray devices. We proceed to demonstrate, however, that the inverse experiment—the spontaneous parametric decay of an x-ray photon (the pump) into an optical photon (the signal) and another x-ray photon (the idler)—is presently feasible. A generically related process, in which both the signal and the idler are in the x-ray region, has been previously discussed by the present authors² and very recently observed experimentally.³

Quantum noise at the signal and idler frequencies (ω_s and ω_i , respectively) produces in the crystal a fluctuating polarization $\vec{\mathcal{P}}_p$, at the pump frequency ($\omega_p = \omega_s + \omega_i$) which, as discussed by Kleinman,⁴ interacts with the input pump field \vec{E}_p via $\int_V d^3r \int {}_{\perp}\vec{\mathcal{P}}_p \cdot d\vec{E}_p$. The nonlinear susceptibility relevant to this process is seen to be that for frequency summation of input fields at ω_s and ω_i to produce an output at ω_p . The integration over the crystal volume requires that momentum be conserved, and since the nonlinear susceptibility is spatially periodic, this requirement may be met by using a reciprocal lattice vector of the crystal, $\vec{Q}(hkl)$, such that

$$\vec{k}_p = \vec{k}_s + \vec{k}_i + \vec{Q}(hkl), \quad (3)$$

where the \vec{k} 's are the propagation vectors of the photon fields. Equation (3) is the law of nonlinear diffraction⁵ as applied to the present problem.

A formal quantum-mechanical expression, convenient to our purposes, for the coherent, nonlinear response of an atom to applied electromagnetic fields has been given by Armstrong et al.⁶ Of the several terms that occur, only one is of importance in the present instance, enabling us to write (under the assumption that the wave functions may be taken as real)

$${}_{\perp}\vec{\mathcal{P}}_p(\omega_p) = \frac{e^3 {}_{\perp}\vec{E}_i}{m^2 \omega_p \omega_i} \text{Re} \left[\exp(-i\omega_p t) \sum_j \left\{ \langle 0 | \exp[i(\vec{k}_i - \vec{k}_p) \cdot \vec{r}] | j \rangle \langle j | \exp(i\vec{k}_s \cdot \vec{r}) \nabla \cdot \vec{E}_s | 0 \rangle \frac{1}{\Omega_j^2 - \omega_s^2} \right\} \right], \quad (4)$$

where Ω_j is the resonant frequency of the j th state, and the other symbols have their usual meaning. Making the dipole approximation with respect to the optical (signal) field, transforming from the matrix elements of ∇ to those of \vec{r} , setting $-\vec{Q} = \vec{\kappa}_i - \vec{\kappa}_p$ (generally, $\kappa_s < 10^{-3}Q$), and writing $\omega_p \cong \omega_i = \omega_x$, $\omega_s = \omega_0$, we observe that Eq. (4) becomes equivalent to Eq. (2) with

$$\delta\rho(\vec{r}, \vec{E}_0) \equiv \delta(e\Psi_0^*\Psi_0) = 2e\Psi_0\delta\Psi_0, \quad (5)$$

where Ψ_0 is the ground-state wave function and $\delta\Psi_0$ is the perturbation correction induced by the optical field:

$$\delta\Psi_0 = \frac{e}{\hbar} \sum_j \left\{ \Psi_j \langle j | \vec{r} \cdot \vec{E}_0 | 0 \rangle \frac{\Omega_j}{\Omega_j^2 - \omega_0^2} \right\}. \quad (6)$$

Depending upon the particular problem at hand, it may frequently be convenient to interpret $\delta\rho$ in terms of some other property of the system. Since we require a numerical estimate of Eq. (4), this is true also in the present instance. We proceed as follows: We associate with $\delta\rho(\vec{r}, \vec{E}_0)$ a polarization density, $\delta\vec{p}(\vec{r}, \vec{E}_0)$, using the continuity equation

$$\delta\rho(\vec{r}, \vec{E}_0) = -\nabla \cdot \delta\vec{p}(\vec{r}, \vec{E}_0), \quad (7)$$

and define an optical polarizability density $\vec{\alpha}_0(\vec{r})$, such that

$$\delta\vec{p}(\vec{r}, \vec{E}_0) \equiv \vec{\alpha}_0(\vec{r}) \cdot \vec{E}_0,$$

whence

$$\delta\rho(\vec{r}, \vec{E}_0) = -\nabla \cdot [\vec{\alpha}_0(\vec{r}) \cdot \vec{E}_0]. \quad (8)$$

Our interpretation of $\vec{\alpha}_0(\vec{r})$ is that $\vec{\alpha}_0(\vec{r})d^3r$ describes the contribution of a small volume of charge at the point \vec{r} to the total optical polarizability $\vec{\alpha}_0$; that is, we require

$$\vec{\alpha}_0 \cdot \vec{E}_0 = \left[\int \vec{\alpha}_0(\vec{r}) d^3r \right] \cdot \vec{E}_0. \quad (9)$$

This condition is, indeed, satisfied, as may be seen by using the exact quantum-mechanical result

$$\vec{\alpha}_0 \cdot \vec{E}_0 = \int \vec{r} \delta\rho(\vec{r}, \vec{E}_0) d^3r. \quad (10)$$

Equation (4) may now be written

$${}_1\vec{\mathcal{P}}_p(\omega_p) = \frac{e}{} \frac{{}_1\vec{E}_i}{2m\omega_x^2} \text{Re} \left\{ i \exp(-i\omega_p t) \vec{Q} \cdot \left[\int e^{-i\vec{Q} \cdot \vec{r}} \vec{\alpha}_0(\vec{r}) d^3r \right] \cdot \vec{E}_0 \right\}. \quad (11)$$

It is clear that the valence electrons will make the dominant contribution to $\vec{\alpha}_0(\vec{r})$. Recently, the optical properties of a wide range of ionic and covalent materials has been very successfully described⁷ by a simple model that ascribes all of the linear (and nonlinear) polarizability of the medium to that part of the valence electron charge density that resides in the chemical bond—the “bond charge.” In view of this we assume for $\vec{\alpha}_0(\vec{r})$

$$\vec{\alpha}_0(\vec{r}) = \alpha_0 \rho_B(\vec{r}) \vec{I}, \quad (12)$$

where $\rho_B(\vec{r})$ is the distribution function for the bond charge and α_0 is the measured optical polarizability. From Eq. (9) we see that the scaling required for $\rho_B(\vec{r})$ is that $\int \rho_B(\vec{r}) d^3r = 1$. Writing ${}_1\vec{\mathcal{P}}_p(\omega_p) = {}_1\vec{g}(hkl) E_s E_i$, we have for the vector nonlinear atomic scattering factor, ${}_1\vec{g}(hkl)$,

$${}_1\vec{g}(hkl) = \frac{ie}{2m\omega_x^2} \alpha_0 \left\{ \int \exp[-i\vec{Q}(hkl) \cdot \vec{r}] \rho_B(\vec{r}) d^3r \right\} \vec{\theta}_{psi}(hkl),$$

where

$$\vec{\theta}_{psi}(hkl) = [\hat{v} \times (\hat{v} \times \hat{a}_i)] [\hat{a}_s \cdot \vec{Q}(hkl)], \quad (13)$$

and \hat{v} is a unit vector along $\vec{\kappa}_p$, while \hat{a}_s, \hat{a}_i are unit vectors along \vec{E}_s and \vec{E}_i , respectively. We observe that Eq. (13) follows from Eq. (4) as an exact quantum-mechanical result for the isotropic, three-dimensional harmonic oscillator, as well as for a two-level system. The limit of Eq. (11) for small \vec{Q} is also easily obtained as an exact result from Eq. (4) for any system. We note, in addition,

the important sum rule

$$\int d\vec{Q} \int d^3r e^{-i\vec{Q}\cdot\vec{r}} \delta\rho(\vec{r}, \vec{E}_0) = 0. \tag{14}$$

The vector nonlinear structure factor $\perp\vec{G}(hkl)$, is, as defined previously²

$$\perp\vec{G}(hkl) = \sum_n \perp\vec{g}_n(hkl) \exp[-2\pi i(hu_n + kv_n + lw_n)] \tag{15}$$

with u_n, v_n, w_n the fractional coordinates of atom n in the unit cell, and the sum is taken over all atoms in the cell. The macroscopic vector nonlinear susceptibility $\perp\chi_{NL}(\omega_x = \omega_0 + \omega_x) = N \perp\vec{G}(hkl)$, where N is the number density of unit cells. The real and imaginary parts of χ_{NL} add in quadrature, and, in addition, $|\chi_{NL}|^2$ must be summed over all interacting modes of polarization of the pump, signal, and idler fields.^{4,8}

If the crystal is oriented at Bragg's angle for diffraction of the pump, the optical signal is emitted on the surface of a cone of revolution whose axis is parallel to the diffracted pump direction, and whose apex angle, $2a$, is determined by $\cos a = 1/n_0$, where n_0 is the refractive index of the medium at the optical frequency. Since the dispersion in n_0 is generally small, a very broad optical spectrum is emitted along a , resulting in a large value for the signal power per unit solid angle. This is an example of the "edge enhancement" described by Kleinman.⁴ In spite of this enhancement, we favor for photoelectric detection a different geometry in which a narrow signal spectrum is emitted into a large solid angle. This occurs when the crystal is oriented as shown in Fig. 1. Writing

$$\varphi(hkl) \equiv \int e^{-i\vec{Q}\cdot\vec{r}} \rho_B(\vec{r}) d^3r \quad (\varphi[000] = 1),$$

restricting ourselves to reflections for which all atoms radiate in phase so that $\perp\vec{G}_x = n \perp\vec{g}$, where n is the number of atoms in the cell, and assuming an unpolarized source of x rays, we may, following Kleinman,⁴ write for the number of signal photons counted each second, \dot{N}_s ,

$$\dot{N}_s \cong \left(\frac{4\pi^3 r_0}{\mu_0} \right) \left(\frac{\lambda_x I_p \Delta\Omega_s}{\lambda_0^3} \right) \left[\frac{n_0 \chi_L^2 \varphi^2(hkl) V}{1 + n_0} \right] \sin^2 2\theta_B (2 - \sin^2 2\theta_B). \tag{16}$$

Here r_0 is the classical radius of the electron, μ_0 is the energy of its rest mass, λ_x and λ_0 are the wavelengths of the source and signal, respectively, χ_L is the measured linear optical susceptibility at the wavelength λ_0 (this automatically includes the proper local-field correction), θ_B is Bragg's angle for diffraction of the pump using the reciprocal lattice vector chosen to satisfy Eq. (3), I_p is the intensity of the pump, and V , in the absence of extinction at the pump frequency and absorption at the signal frequency, is given by

$$V = \frac{1}{(2\pi)^3} \int d^3\kappa_i \int_{-\infty}^{\infty} \mathcal{V}(\vec{r}) \exp\{-i[\vec{\kappa}_p - \vec{\kappa}_s - \vec{\kappa}_i - \vec{Q}(hkl)] \cdot \vec{r}\} d^3r, \tag{17}$$

where $\mathcal{V}(\vec{r})$ is the volume common to the sample and a replica displaced by \vec{r} , and where the integration on $\vec{\kappa}_i$ is over the bandwidth and angular aperture of the signal detector. If these limits are sufficiently great (for the experiments described here this will, of necessity, almost always be true), then the integration may be extended over all κ_i space, and we retrieve the important result that $V - \mathcal{V}(0)$ equals the illuminated sample volume, independent of shape or orientation. This remains true in the presence of extinction at the pump (and idler⁹) frequencies, and also for an imperfect crystal; these effects simply serve to broaden the optical spectrum. When thermal effects are important, Eq. (17) must be multiplied by the usual Debye-Waller temperature factor. Since, at least initially, the experiment will be count-rate limited, we assume that the sample is large enough to intercept all of the available pump, so that

$$I_p V = P_p / \mu_p, \tag{18}$$

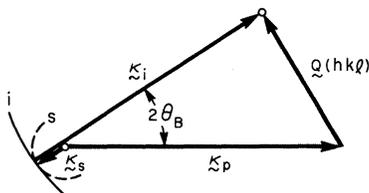


FIG. 1. Wave vector matching diagram. The geometry shown yields a narrow signal spectrum into a large solid angle because the signal (s) and idler (i) surfaces osculate.

where P_p is the total pump power delivered to the sample, and μ_p is the total extinction coefficient at λ_p . When Eq. (18) is applicable, the possibility of precision measurements of $\delta\rho$ exists because the usual extinction corrections that plague ordinary Bragg diffraction are unnecessary.

If $\Delta/2$ is the deviation of the signal direction from collinearity with the idler, as in Fig. 1, then the optical wavelength changes as

$$\Delta\lambda_0/\lambda_0 \approx [1 - \cos(\Delta/2)]/[1 + (1/n_0) \cos(\Delta/2)]. \quad (19)$$

For $\Delta = 30^\circ$, and $n_0 \approx 2.5$, $\Delta\lambda/\lambda_0$ is seen to be only $\sim 2.5\%$. The major source of signal spectrum broadening will be due not to the large collection angle, but rather to the pump divergence. If the angular divergence of the pump in the plane of Fig. 1(a) is δ_I , and out of the plane is δ_0 , and if the pump width is $\Delta\lambda_x$, then

$$\delta_I \approx \frac{n_0 + 1}{\sin 2\theta_B} \left(\frac{\Delta\lambda_0}{\lambda_0} \right) \left(\frac{\lambda_x}{\lambda_0} \right), \quad \delta_0^2 \approx 4 \frac{n_0 + 1}{\sin^2 \theta_B} \left(\frac{\Delta\lambda_0}{\lambda_0} \right) \left(\frac{\lambda_x}{\lambda_0} \right), \quad (20)$$

and

$$\frac{\Delta\lambda_0}{\lambda_0} \approx \frac{\sin^2 \theta_B}{n_0 + 1} \left(\frac{\lambda_0}{\lambda_x} \right) \left(\frac{\Delta\lambda_x}{\lambda_x} \right).$$

The effect of the crystal imperfection is similar to that of the in-plane pump divergence, with δ_I being now the width of the rocking curve.

We estimate \dot{N}_s [Eq. (16)] for the (111) reflection of diamond. We choose $\Delta\lambda_0/\lambda_0 \approx 20\%$, $\lambda_0 = 3500 \text{ \AA}$ (this region is adequately free of fluorescence in type II diamonds), $\Delta\Omega_s = 0.274 \text{ sr}$ ($\Delta = 30^\circ$), and $\lambda_x = 1.54 \text{ \AA}$ (Cu $K\alpha$). We calculate that with a fine-focus x-ray source and suitable optics, it is presently feasible to deliver to the sample, within the rather stringent limits imposed by Eq. (20), a flux of $10 \mu\text{W/kW}$ of electrical power dissipated at the anode. Suitable 30-kW sources are now offered commercially, so that we assume $P_p = 300 \mu\text{W}$. We approximate the contribution of the l th C-C bond to $\rho_B(\vec{r})$ by a normalized spherical Gaussian centered at \vec{r}_l , the midpoint of the bond, so that

$$\rho_B(\vec{r}) = \frac{1}{4} \left(\frac{\sigma}{\sqrt{\pi}} \right)^3 \sum_{i=1}^4 \exp[-(\vec{r} - \vec{r}_i)^2 \sigma^2] \text{ and } \varphi(hkl) = \frac{1}{4} e^{-(Q/2\sigma)^2} \sum_{i=1}^4 \exp(-i\vec{Q} \cdot \vec{r}_i) \quad (21)$$

with the sum over all four covalent bonds. From the work of Dawson¹⁰ we estimate $\sigma \approx 2.0 \text{ \AA}^{-1}$. With these approximations we obtain $\dot{N}_s \approx 5/\text{sec}$. An enormous discrimination against fluorescence and other sources of noise may be obtained by simultaneously detecting in fast time coincidence both the optical signal and x-ray idler. This is feasible because the idler is emitted in a very small solid angle, of order 10^{-5} sr or less, so that x-ray fluorescence and Compton scattering are not serious problems. Several important aspects of such a scheme have been discussed by the present authors¹¹ for the case where both the signal and idler are in the x-ray region; the principles enumerated there are also applicable in the present instance.

If the optical polarizability is known, an absolute value of $\delta\rho/E_0$ can be obtained from a series of relative measurements of intensity and the use of Eq. (10). The usual phase uncertainty always present in diffraction experiments exists here too, but since the crystal structure may be considered known, the use of simple models for a starting point, together with the sum rule given in Eq. (14) and the knowledge that the integral of $\delta\rho(\vec{r})$ vanishes, should permit a complete solution. There are, of course, many ways of modulating the electronic charge density and we anticipate that some of these may prove useful in experiments similar to the one described here. The special advantage of an optical-frequency modulation is that the nuclei are effectively clamped. In any event, we may expect that the knowledge of $\delta\rho(\vec{r}, \vec{E}_0)$ will prove to be useful in the interpretation of many phenomena in atomic and solid state physics. As an example, we present a form for the nonlinear optical polarizability, $\beta_{xyz}^{\omega_3 = \omega_1 + \omega_2}$. Writing $\mathfrak{D}_x^{\omega_i}(\vec{r}) \equiv \partial[\delta\rho(\vec{r}, \vec{E}(\omega_i))]/\partial E_x$, etc., we find as an excellent approximation which includes the important aspects of dispersion that

$$\beta_{xyz}^{\omega_3 = \omega_1 + \omega_2} \approx \frac{1}{4} \int \frac{d^3r}{\rho(\vec{r})} \{x \mathfrak{D}_y^{\omega_1}(\vec{r}) \mathfrak{D}_z^{\omega_2}(\vec{r}) + y \mathfrak{D}_x^{\omega_3}(\vec{r}) \mathfrak{D}_z^{\omega_2}(\vec{r}) + z \mathfrak{D}_x^{\omega_3}(\vec{r}) \mathfrak{D}_y^{\omega_1}(\vec{r})\}. \quad (22)$$

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Relativistic Magnetic Dipole Emission: Lifetime of the $1s2s\ ^3S_1$ State of Heliumlike Argon*

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The lifetime of the $1s2s\ ^3S_1$ state of the heliumlike atom Ar XVII has been measured by observing the decay in flight of the metastable component of a fast foil-excited beam. The decay occurs predominantly by relativistic magnetic-dipole emission, a process first discussed by Breit and Teller. The result, $\tau(2^3S_1) = (172 \pm 30) \times 10^{-9}$ sec, is compared with a recent calculation by Drake of the $M1$ transition probability.

Until recently it was believed that the primary decay mode for the $1s2s\ ^3S_1$ state of two-electron atoms would be spin-orbit-induced double electric-dipole ($2E1$) emission. This process was first suggested by Breit and Teller,¹ and has been accurately calculated by Drake, Victor, and Dalgarno² and Bely and Faucher.³ However, in 1969 Gabriel and Jordan⁴ reported the observation of solar coronal lines corresponding to the $1s2s\ ^3S_1-1s^2\ ^1S_0$ energy separation for the helium isoelectronic sequence C V-Mg XI, and the Fe XXV line has been reported by Neupert and Swartz,⁵ indicating that the primary decay mode is single-photon emission. Single-photon emission also was discussed by Breit and Teller,¹ who pointed out that relativistic effects can lead to magnetic-dipole radiation (this is identically zero in the nonrelativistic approximation). More recently Schwartz⁶ and Drake⁷ have studied this process and conclude that, to relative accuracy Z^{-1} , the dominant contributions come from kinetic-energy and finite-wavelength corrections to the magnetic-dipole moment, which have non-zero matrix elements between the 1^1S_0 and 2^3S_1 states. Schwartz⁶ has calculated the rate of this process using hydrogenic wave functions and energies. He obtains the asymptotic (to large Z) result $A_{M1}(2^3S_1-1^1S_0) = 1.66 \times 10^{-6} Z^{10}$ sec⁻¹, which yields, for argon ($Z = 18$), $\tau_{M1}(2^3S_1) = 169$ nsec.

More accurate calculations have been performed by Drake⁷ using correlated wave functions and energies, with the result for argon $\tau_{M1}(2^3S_1) = 194$ nsec.

The astrophysical importance of the lifetime of the 2^3S_1 state has recently been emphasized by Gabriel and Jordan.⁸ These authors have developed a theory for deducing the electron density in the solar corona based on intensity measurements. A crucial parameter in this theory is the 2^3S_1 lifetime and they have derived a semi-empirical value⁹ of $\tau_{M1}(2^3S_1) = 2.3 \times 10^{-10} \lambda^5$ sec (λ in Å) which yields for argon a value of 230 nsec. The $2^3S_1-1^1S_0$ transition in the heliumlike ions Si XIII, S XV, and Ar XVII has been observed in the laboratory by Marrus and Schmieder¹⁰ with the beam-foil method, thus confirming the single-photon decay mode, but as yet no experimental lifetime of any $1s2s\ ^3S_1$ state has been reported. In this Letter we report the measurement of the lifetime of the $1s2s\ ^3S_1$ state of Ar XVII, using the beam-foil method. The result is

$$\tau(2^3S_1) = 172 \pm 30 \text{ nsec.}$$

The apparatus used in this measurement is illustrated in Fig. 1. Argon-40 ions in the +14 charge state having an energy of 10.3 MeV/nucleon ($\beta = 0.148$) are obtained from the Berkeley heavy-ion linear accelerator (HILAC) and are