Second-harmonic photons from the interaction of free electrons with intense laser radiation

T. J. Englert and E. A. Rinehart
Department of Physics and Astronomy, University of Wyoming, Laramie, Wyoming 82071
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A preliminary experimental investigation of the generation of second-harmonic photons from the interaction of free electrons with an intense laser beam has been performed. Second-harmonic photons with a wavelength of 530 nm generated from the interaction of free electrons with 1060-nm photons from a neodymium-glass laser are implied by observing Doppler-shifted photons with wavelengths of 490 and 507 nm. The observed photon wavelengths result from a Doppler shift of the laser photon wavelength as viewed in the rest frame of the electrons, combined with a Doppler shift of the second-harmonic photon emitted from 1600- and 500-eV electrons. Comparison of experimental results with those predicted by cross sections, derived from classical and quantum electrodynamics, shows reasonable agreement with both theories. Although second-harmonic photons are created, the dynamics of second-harmonic-photon generation (accelerated electron motion due to the electromagnetic field or actual two-photon interaction with the electron) cannot be resolved without further experiment.

I. INTRODUCTION

Since the numerically successful explanation of the Lamb shift by quantum electrodynamics (QED), many of the experimental efforts directed at verification of QED have dealt with measurements of small energy differences of bound states in atoms or atomiclike systems.[1–3] Conceptually, the QED treatment of such systems, if taken literally, relies upon physically unseemly processes of vacuum fluctuations and virtual-photon interaction with charged particles leading to radiative corrections of these bound-state energy levels. We would expect that such a theory, whose foundations are rooted in the quantization of electromagnetic fields, must surely provide equally accurate descriptions of the interaction of charged particles with fields made up of real photons. The single-photon processes of Thomson and Compton scattering are in fact known to be accurately described by QED.[4–6]

The QED description of the interaction of free electrons with the photons of an intense radiation field leads to the prediction of multiphoton processes including such phenomena as stimulated Compton scattering and harmonic-photon generation, which one would assume should both be observable. Measurements of stimulated Compton scattering have been made and agree with the predictions of QED[7]; however, relatively few experiments have been performed which deal with multiphoton interaction with free electrons.

Of particular interest is the process of second-harmonic-photon generation, predicted by classical approaches as well as QED.[8–11] The cross sections, calculated by classical and QED techniques, for generation of second-harmonic photons from free electrons in an intense radiation field, are in relatively good agreement with one another; however, the interpretation of the dynamical processes involved are quite different. Classically, the second-harmonic radiation results from anharmonic acceleration of the electron as it interacts with the radiation field, while quantum mechanically, the second-harmonic photon may be interpreted as resulting from the interaction of two photons with the electron, the energy of the second-harmonic photon being the sum of the energies of the two incident photons. In QED treatments of such a process one often sees the interaction represented as $\gamma_1+\gamma_1+e\rightarrow\gamma_2+e'$. The latter dynamical process is certainly very intriguing since an apparent absorption of two photons, by an electron, followed by the emission of a single, energy-conserving photon, may be indicative of electron structure. Perhaps either of these interpretations (classical or quantum) is too literal and naive due to our ignorance of the nature of photons and electrons. This ignorance is undoubtedly already manifest in runaway solutions, preaccelerations, and infinite renormalization processes, to name but a few of the problems that beset the theories of electrodynamics. Such problems have prompted a revivial of attempts to describe the electron as an extended particle.[12–14]

The cross section for harmonic radiation generated from free-electron interaction with polarized light, obtained by Vachaspati[8] using a classical approach, is found to be

$$(d\sigma_H/d\Omega)_{\text{class}}=\tfrac{1}{16}(e^2/mc^2)^2(2e^2I_0\lambda^2/m^2c^5)\times(9\sin^2 2\alpha-24\cos^2\alpha\cos\theta+4\sin^2\theta)\ , \quad (1)$$

where the angular dependence is such that $\cos\theta=\hat{n}\cdot\hat{n}_0$ and $\cos\alpha=\hat{n}\cdot\hat{e}_0$ with $\hat{e}_0=\vec{E}_0/E_0$, $\hat{n}_0=\vec{k}_0/k_0$, and the direction of observed scattered radiation given by $\hat{n}$. I_0 and λ are, respectively, the intensity and wavelength of the radiation incident on the electron.

Using QED, the cross section found by Jafarpour,[11] essentially in agreement with other authors,[4,9,10] except for minor differences in angular dependence, is given by

$$\left[\frac{d\sigma_H}{d\Omega}\right]_{\text{QED}}=\frac{4\pi e^2 I_0 B^2}{m^2c^5K^2}\frac{d\sigma_T}{d\Omega}\ , \tag{2}$$

where

$$B=[\hat{K}\cdot\hat{e}(\vec{K})+2\hat{k}\cdot\hat{e}(\vec{K})\hat{e}(\vec{K})\cdot\hat{e}(\vec{k})]/\hat{e}(\vec{K})\cdot\hat{e}(\vec{k})\ . \tag{3}$$

Here $\hat{K}$ and $\hat{k}$ are the respective directions of intense radiation and scattered photon propagation, $\hat{e}(\vec{K})$ and $\hat{e}(\vec{k})$ are the corresponding polarization vectors, and $K=2\pi/\lambda$. The factor $d\sigma_T/d\Omega$ is the Thomson cross section. It is frequently argued that the absence of Planck's constant in the cross section found by QED techniques [Eq. (2)] suggests that the process of second-harmonic—radiation generation from the interaction of free electrons with an intense electromagnetic field does not depend upon quantization of the field. Such arguments may be supported in the classical derivation by Vachaspati; however, the quantum characteristics of the field remains controversial.[15] As a first step in a possible resolution of the question of the dynamics involved, it is imperative that we determine whether or not second-harmonic photons do, in fact, result from the interaction of free electrons with intense light. In this paper we report on an experimental observation of harmonic photons generated by the interaction of laser radiation with free electrons and obtain reasonable agreement with the predicted cross sections for the process.

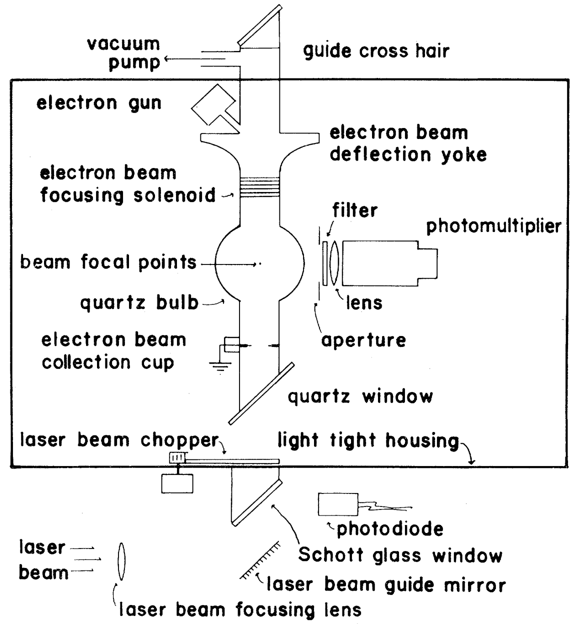

FIG. 1. Details of the experimental arrangement (not to scale). Overall length of the scattering chamber is approximately 70 cm. Diameter of the bulb at the beam focal points is 8 cm.

II. EXPERIMENTAL PROCEDURE

The focused beam from a pulsed neodymium-glass laser collides head on with a continuous, focused, low-energy beam of electrons. The electron source and focusing is described elsewhere.[16] Maximum focusing of the two beams occurs at approximately the same position, each beam having a radius of 0.1 ± 0.01 mm, in order to simultaneously achieve maximum photon and electron densities at the point of observation. Since the wavelength of the laser radiation is 1060 nm, the second-harmonic photons have a convenient (for detection purposes) wavelength of approximately 530 nm. The laser is allowed to oscillate freely in all permissible cavity modes giving a total energy output of 1.08 ± 0.15 J per laser firing. An average of 20 pulses, on the order of 10^{-12} sec duration each, of laser radiation per laser firing occurs, giving a mean value of approximately 0.05 J per laser pulse. Given the foregoing parameters, a laser-beam intensity of about 1.7×10^{14} W/cm^2 is obtained. It is of interest to note that given this intensity and a laser-photon wavelength of 1060 nm, a simple calculation due to Eberly[17] shows we can expect an average of about 2.4 photons to be in the vicinity of an electron at any given time during a laser-beam pulse. Electron-beam energies of 500 and 1600 eV are used in the experiment with a nominal beam current of 2×10^{-3} A. This provides electron densities on the order of $10^{10}/cm^3$. The electron-beam energies will be discussed later. Figure 1 shows the experimental layout.

Detection of photons having wavelengths harmonic to the laser radiation is accomplished by placing an interference filter between the interaction site, defined by the colliding beams, and the photomultiplier. Since the light transmitted by interference filters is dependent upon the angle of incidence, an aperture is used to discriminate against photons with wavelengths much different from the harmonic-photon wavelength, from entering the photomultiplier. A simple lens is also included to slightly refocus the diverging light from the interaction site onto the active area of the photocathode. Laser light pulses are detected by a photodiode placed in that part of the laser beam scattered from the Schott glass entry window of the housing of the experimental apparatus. Temporal coincidence to within 10^{-7} sec is then required between laser and photomultiplier pulses. Photodiode pulses were electronically delayed approximately 1.24×10^{-6} sec to compensate for pulse processing in the photomultiplier leg of the counting apparatus. A block diagram of the counting and coincidence arrangement is shown in Fig. 2.

Pressures within the scattering chamber during the experiment range from 2×10^{-6} to 5×10^{-7} Torr, leaving on the order of 10^9 residual gas particles per cubic centimeter in the chamber. Extraneous radiation due to laser-beam interaction with the residual gases (e.g., multiphoton processes), as well as light from the electron beam (bremstrahlung, etc.), will be expected to contribute to the background. It is essential that we determine as nearly as possible that the observed events attributed to second-harmonic-photon generation are associated with the electrons themselves and that the backgrounds due to other processes are adequately accounted for.

In an effort to ensure that the source of observed

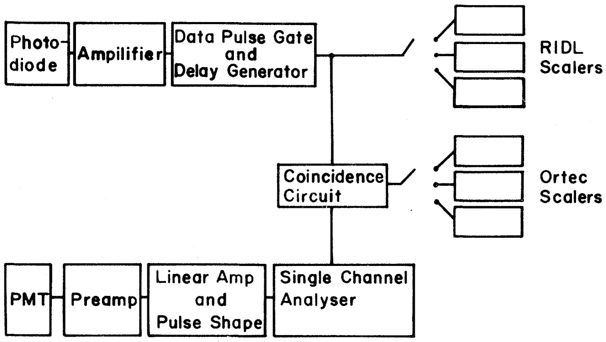

FIG. 2. Block diagram of counting and coincidence arrangement.

second-harmonic photons is indeed the electron beam, two different electron-beam energies of 500 and 1600 eV are used. The resulting Doppler-shifted second-harmonic photons from the laser-beam interaction with 500- and 1600-eV electrons are calculated to have wavelengths λ_H of 507 and 491 nm, respectively, when emitted in a direction perpendicular to the electron motion. A summary of the Doppler-shift calculations may be found in Table I. By using 510- and 490-nm interference filters, each having 10-nm bandpass, we determine whether the observed second-harmonic-photon events come and go according to electron-beam energy and appropriate passing or blocking filters. The interference filters used were originally designed as 500- and 520-nm filters, however, sufficient electron-beam currents were unobtainable at the low accelerating voltage required to yield 520-nm harmonic photons. More convenient operation was achieved by rotating the filters through an approximate angle of 22°, thus providing a shift of about 10 nm in the transmission peaks. Filter transmission as a function of incident radiation wavelength is shown in Fig. 3.

Three modes of data collection are used.

L-e mode. The laser and electron beams are allowed to collide.

L-$\bar{e}$ mode. The electron beam is turned off for approximately 2 sec centered around the time of laser firing. The laser beam propagates through the scattering chamber as in the L-e mode.

$\bar{L}$-e mode. The laser is allowed to fire but is mechanically chopped out of the experiment just before entering the scattering chamber. The electron beam propagates through the scattering chamber as in the L-e mode. Each run then consists of three sets of data accumulated in the modes described. Automatic switching after each laser firing (one per minute) provides for sequential alternation of the L-e, L-$\bar{e}$, and $\bar{L}$-e modes. Coincidences between photon events and laser pulses are counted in each mode, as are the number of laser pulses. Data are recorded as coincident events per laser pulse in each of the modes of data collection, and the net events per laser pulse N/P_L is then found according to

$$N/P_L=(C/P_L)_{L\text{-}e}-(C/P_L)_{L\text{-}\bar{e}}-(C/P_L)_{\bar{L}\text{-}e}\ , \tag{4}$$

where C is the number of coincidence pulses accumulated during the run, and P_L is the number of laser pulses. The

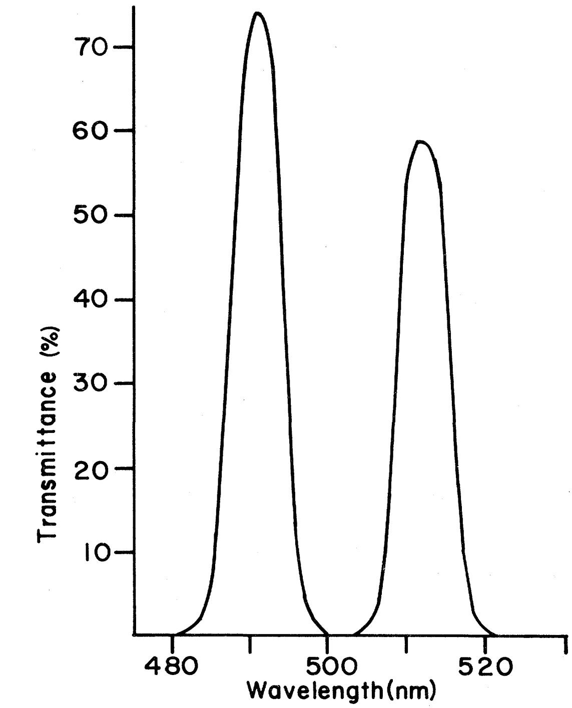

FIG. 3. Transmissions as a function of wavelength after rotation of 500- and 520-nm interference filters through approximately 22°.

TABLE I. Summary of the calculations of the Doppler-shifted second-harmonic-photon wavelengths. The laser radiation has a wavelength of 1060 nm, as measured in the laboratory, and collides head on with the electron beam. Second-harmonic photons are detected at right angles ($\theta=\pi/2$) to the direction of the colliding beams. Primes indicate the electron rest frame, while unprimed symbols indicate laboratory frame values.

Electron energy (eV)	$\beta=v/c$	$\lambda'=\lambda\left[\frac{1-\beta}{1+\beta}\right]^{1/2}$ (nm)	$\lambda'_H=\frac{\lambda'}{2}$ (nm)	$\lambda_H=\lambda'_H\frac{1-\beta\cos\theta}{(1-\beta^2)^{1/2}}$ (nm)
500	0.0442	1014	507	507
1600	0.0789	979	489.5	491

average signal-to-noise ratio is 2.1:1.

As nearly as possible a complete set of runs was obtained by a sequence of data accumulations as follows.

(a) For 500-eV electrons, data were accumulated using the 510-nm filter, followed by an accumulation using the 490-nm filter, followed by a repeated accumulation using the 510-nm filter.

(b) For 1600-eV electrons, data were accumulated using the 490-nm filter, followed by an accumulation using the 510-nm filter, followed by a repeated accumulation using the 490-nm filter.

This procedure should lend some assurance that the integrity of the beams is maintained during data accumulation and that data taken with the pass filter and blocking filter are consistent. Times required for each run are on the order of 3 h. In the event that a sequence was interrupted, those data accumulated up to the time of interruption were retained, thus giving a somewhat higher than 2:1 ratio of pass-filter data compared to blocking-filter data.

III. EXPERIMENTAL RESULTS

The histograms of Fig. 4 show the distributions of runs with yields of net second-harmonic photons per laser pulse according to electron-beam energies of 500 and 1600 eV and respective passing and blocking filters. Net second-harmonic photons are determined by Eq. (4). These histograms are not normalized for differences in filter transmission. Table II gives the respective yields of net second-harmonic photons at these electron energies with the quoted errors reflecting a least-squares weighted average based on Poisson statistics in each case. Figure 5 shows a compilation of all data normalized for filter transmission. The variation about the mean of harmonic photons per laser pulse indicated in the histograms of Figs. 4 and 5 is seen to exceed the calculated errors, due to counting statistics, by nearly a factor of 10. This apparent excessive fluctuation in the generation of these second-harmonic photons may be closely related to similar fluctuations reported in earlier work done with harmonic generation.[18,19] It has been shown that when lasers operating in more than a single mode are used in the generation of second harmonics in a medium, considerable statistical fluctuation in the ratio of harmonic intensity to fundamental intensity results. We have already noted that the output from the laser used in the current experiment contains several modes whose phase relationships are very likely random (no attempts of measurement of phase relationship have been made in the present work), and so we might anticipate that intensity fluctuations in the second-harmonic photons, due to the presence of several modes in the fundamental beam, may be manifest in the work described here. We might also attribute some of the fluctuation to uncertainty in beam alignment since such alignment was accomplished rather crudely by simply watching for a net increase in harmonic photons while carefully adjusting the laser-beam guide mirror. When more careful measurements are made in future experiments of this nature, we must surely give closer attention to the details of effects of multimode laser output.

As a further indication that the source of detected harmonic photons is the electrons, a measure of net harmonic photons per laser pulse as a function of electron-beam current has been made (see Fig. 6). It would be expected that the number of harmonic photons should be directly proportional to the number of electrons present as seen in Fig. 6.

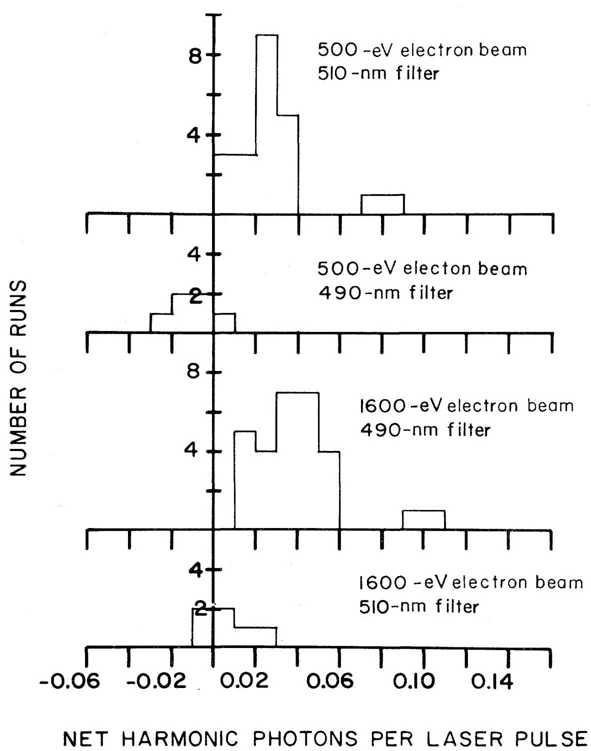

FIG. 4. Distribution of runs showing net second-harmonic photons per laser pulse [see Eq. (4)] from the interaction of free electrons with intense laser radiation for a laser-beam intensity of 1.7×10^{14} W/cm^2 and electron density of approximately $10^{10}/cm^3$. Detector subtends only 0.024 of the total solid angle.

TABLE II. Net yield of Doppler-shifted second-harmonic photons per laser pulse from the interaction of free electrons with laser radiation. Laser radiation with an intensity of 1.7×10^{14} W/cm^2 and wavelength of 1060 nm collides with an electron beam having an electron density of approximately $10^{10}/cm^3$. The detector subtends 0.024 of the total solid angle at the interaction site. All data are normalized to 74% filter transmission, and errors reflect a least-squares weighted average.

Electron energy (eV)	Filter (nm)	Net harmonic photons per laser pulse
500	510	0.0305 ± 0.0044
500	490	-0.0078 ± 0.0080
1600	490	0.0315 ± 0.0029
1600	510	0.0043 ± 0.0068
Combined data	Passing	0.0312 ± 0.0024
Combined data	Blocking	-0.0016 ± 0.0049

TABLE III. Expected second-harmonic-photon events per laser pulse based on cross sections of Eqs. (4) and (5) and given experimental parameters. Errors reflect uncertainty in experimental parameters.

Cross section used	Calculated total cross section (cm^2)	Expected second-harmonic photons per laser pulse
$(\sigma_H)_{\text{class}}$	3.56×10^{-29}	0.02 ± 0.01
$(\sigma_H)_{\text{QED}}$	4.75×10^{-29}	0.03 ± 0.01

Since some of the experimental methods used here may be considered rather crude, the results of this report should be interpreted as preliminary rather than a precise verification of the cross section for the process of second-harmonic-photon generation due to the interaction of free electrons with intense radiation. In comparing these results with theory, we have, therefore, not attempted an exact integration over the solid angle subtended by the detector, but instead have simply taken $\alpha=0$ and $\theta=\pi/2$ in Eq. (1), giving a total cross section

$$\sigma_H \Big|_{\text{class}}^{\text{pol}} \cong \left[\frac{3}{4\pi}\right](I_0\lambda^2e^2/m^2c^5)\sigma_T \ . \qquad (5)$$

The factor $(e^2/mc^2)^2$ has been written in terms of the total Thomson cross section $\sigma_T=(8\pi/3)(e^2/mc^2)^2$. In evaluation of Eq. (2) to obtain an approximate total cross section as given by QED, we have taken the angular-dependence term [Eq. (3)] to be of order 1 (no attempts were made to determine the polarization of the second-harmonic photons) and proceed in a similar manner to find

$$\sigma_H\mid_{\text{QED}}\cong(4\pi I_0e^2/m^2c^5K^2)\sigma_T=(I_0\lambda^2e^2/\pi m^2c^5)\sigma_T \ . \qquad (6)$$

The fraction of total solid angle subtended by the detector is only 0.024. This value coupled with a total detection efficiency of 10%, including 17% photon detection efficiency of the photomultiplier, 74% transmission of the filter (all data normalized to this value), and a transmission of 80% through the scattering chamber and lens, along with the beam parameters given earlier, yield the predicted second-harmonic photons per laser pulse given in Table III. The errors quoted in Table III reflect an approximation of the uncertainties in experimental parameters used in the calculations.

IV. CONCLUSIONS AND DISCUSSION

Although the results of the experiment described here may only be taken as an order-of-magnitude verification of the cross section for second-harmonic-photon generation from the interaction of free electrons with intense radiation, it leaves little doubt that such a process does occur. The similarities in the theoretical results will surely require more careful evaluation of the cross sections along with more careful experimental testing in order to make the choice of theories clear. It is possible, and perhaps

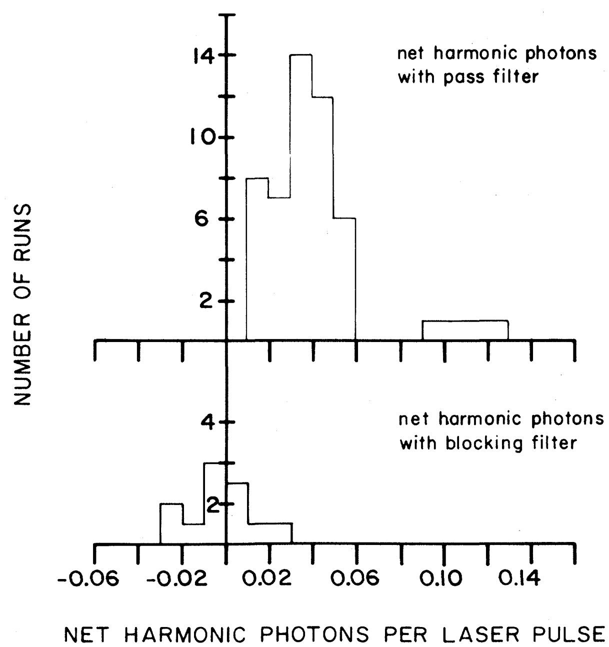

FIG. 5. Distribution of runs showing net second-harmonic photons per laser pulse for combined data from 500- and 1600-eV electrons interacting with an intense laser beam. Laser-beam intensity is 1.7×10^{14} W/cm^2 with a wavelength of 1060 nm. Electron density is approximately 10^{10}/cm^3 and detector subtends only 0.024 of the total solid angle. All data are normalized to 74% filter transmission.

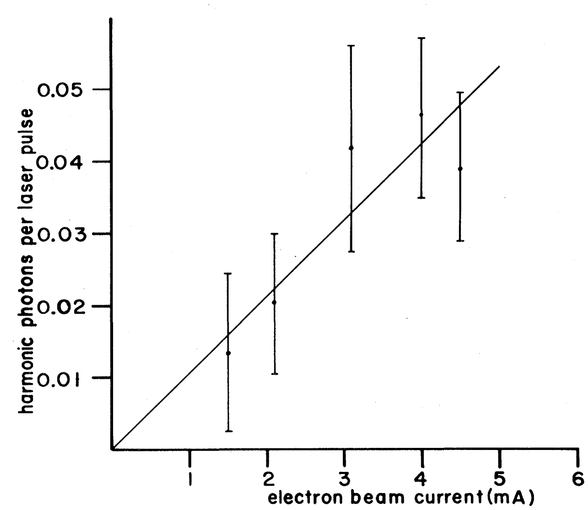

FIG. 6. Dependence of net second-harmonic-photon events per laser pulse on electron-beam current. Solid curve is a linear-regression fit.

likely, that additional attention to these details may still not indicate favor for either the quantum- or classical-field interpretations of the interaction.

The more fundamental question regarding the dynamical processes involved remains debatable. As pointed out by Jaynes,[20] although there are many facets of QED which possess some truths, there are also "elements of nonsense" within the theory. A brief search of the literature published over the past two decades shows considerable theoretical effort directed at processes of multiphoton interaction with free electrons leading to higher harmonics of emitted photons, as well as emitted photons with beat frequencies when the constituents of the incident radiation contain more than one wavelength. It seems essential that experimental tests of a more fundamental nature should be attempted wherein we might obtain more direct information regarding the nature of the dynamics involved.

As emphasized earlier, the work reported here is not to be interpreted as a verification of the dynamics of the interaction of radiation with free electrons, but rather a demonstration that perhaps relatively inexpensive, low-energy experimental tests may be conducted which might conceivably answer questions that are of a more fundamental nature to the understanding of the electron and electrodynamic interactions.

Some insight regarding the quantization of the field and its interaction with a free electron might be gained by investigating the angular momentum involved. If we assume for the moment that the harmonic photon is emitted as a consequence of two photons simultaneously interacting with the electron, the effect during the short time of interaction would be to have a total maximum angular momentum $5\hbar/2$ (assuming the photon spins and electron spin are parallel) associated with the system of the electron and the two photons. The subsequent emission of a single photon will carry away only $1\hbar$ of angular momentum. Such a sequence of events will, at most, result in a final angular momentum of $3\hbar/2$, and angular momentum will not, therefore, be conserved. If, on the other hand, the combination of spins of the two incident photons and the electron yields a total angular momentum of $3\hbar/2$, one can envision that the angular momentum will be conserved with the emission of the harmonic photon, provided the final spin orientation of the electron is flipped. A similar argument may be made for the case where the total angular momentum is $\hbar/2$.

We note in each instance where the sequence of events can be arranged so as to adhere to the conservation of angular momentum, a reversal of the electron-spin results. Furthermore, the inability to conserve angular momentum during second-harmonic photon emission from an initial angular momentum state of $5\hbar/2$ suggests that such interactions are not allowed. A direct measurement of the total spin of the system of two photons and an electron would be extremely interesting and informative, albeit difficult. A more plausible experiment would involve the determination of the dependence of final electron-spin polarization upon initial spin and helicity states of the electrons and photons. It is not certain, without further theoretical consideration, that the observation of such a spin flip would provide a conclusive answer to the question regarding the nature of the interaction, even though spin effects are usually assumed to be inherently quantum mechanical. On the other hand, if it could be verified that the angular momentum states involved required integral units of angular momentum for the field, this would seem to imply some form of field quantization.

It is well known that the point-particle treatment of the electron predicts that the spin of the electron will never be different from $S=\hbar/2$[21]. It is also well known that the point-particle treatment of the electron leads to uncomfortable infinities which have thus far been dealt with through renormalization processes. Suppose, in fact, the electron does not occupy a point, which is certainly a reasonable assumption. It would seem therefore possible that the structured electron would be susceptible to excitations in much the same way that we find all other structured particles to be susceptible to excitation. Assuming that a second-harmonic photon does result from a two-photon interaction with a free electron, we might speculate that such a process hints at electron excitation and, so, some kind of electromagnetic structure of the electron.

There is further speculation that experimental studies of multiphoton interactions with free electrons might also prove to be useful in the investigation of electron structure. Careful determination of the angular dependence for harmonic-photon emission from the interaction of intense radiation with polarized electrons could conceivably provide electromagnetic form-factor data and thus lend further insight into the extended particle concept of the electron.[22]

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