[4]That the large value of $\sigma(e^+e^- \to \text{hadrons})$ for $s \gtrsim 16$ GeV^2 may be due to charmed hadron production was suggested by K. G. Wilson (discussion remarks at Proceedings of the Seventeenth Conference on High Energy Physics, London, 1–10 July 1974), and by R. Shrock and F. Wilczek, unpublished.

[5]J. D. Bjorken and S. L. Glashow, Phys. Lett. 11, 255 (1964).

[6]As this manuscript was being prepared for publication, we learned of a forthcoming paper on the same topic by T. Appelquist, A. De Rújula, D. H. Politzer, and S. L. Glashow, this issue [Phys. Rev. Lett. 34, 365 (1975)].

[7]S. Weinberg, Phys. Rev. Lett. 31, 494 (1973); A. Casher, J. Kogut, and L. Susskind, Phys. Rev. Lett. 31, 792 (1973); K. G. Wilson, Phys. Rev. D 10, 2445 (1974).

[8]J. D. Jackson, private communication; D. R. Yennie, Cornell Univ. Report No. CLNS 291 (to be published).

[9]That $\psi(3695)$ was not observed by J. J. Aubert *et al.* [Phys. Rev. Lett. 33, 1624 (1974)] can be understood as a consequence of two small factors. The first is $\Gamma_e(3.7)/\Gamma_{tot}(3.7) \approx 0.14[\Gamma_e(3.1)/\Gamma_{tot}(3.1)]$. The second refers to the production which can be estimated with the Drell-Yan model. This gives a ratio of 0.10 for the production cross sections.

[10]The $2S$ state is raised to a higher energy than the P state since the former has a node.

[11]For S states $|\psi(0)|^2 = (m_c/4\pi)\langle dV/dr\rangle$; therefore $\eta = 1$ for a linear potential.

[12]We thank K. G. Wilson for providing us with this program.

[13]A purely linear potential gives virtually the same level scheme as our "fit." (The only significant change is that m_c shifts to 2.0 GeV.) This is only an acceptable model if one ignores (Ref. 11) the present indication that $\eta \neq 1$. Thus a precise measurement of η is important. From a theoretical standpoint the Coulomb force should dominate at distances $\lesssim m_c^{-1}$ if the model is to explain the small values of Γ_{tot}.

[14]P-F mixing is negligible.

[15]Should $\psi(3695)$ be $\psi(\text{"}D_1\text{"})$, our numerical predictions would require some change, but the general features shown in Fig. 1 and Tables I and II would survive.

[16]J. D. Jackson, private communication; S. Rudaz, Cornell Univ. Report No. CLNS-293 (to be published).

[17]Direct hadron decays are discussed in Ref. 3.

[18]E. L. Berger, private communication.

[19]H. Braun *et al.*, Phys. Rev. D 2, 488 (1970); G. P. Yost *et al.*, Phys. Rev. D 3, 642 (1971).

[20]J.-E. Augustin *et al.*, SLAC Report No. SLAC-PUB 1520-LBL-3621 (to be published).

COMMENTS

Resonant X-Ray Raman Scattering

Yigal B. Bannett and Isaac Freund
Department of Physics, Bar-Ilan University, Ramat-Gan, Israel
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A theory of the resonant x-ray Raman effect is presented and compared with recent experimental data of Sparks. Excellent agreement between theory and experiment is found for the integrated intensity of the scattering, the spectral density, and the output polarization. The potential importance of this newly discovered spectroscopic probe is discussed.

Although the x-ray Raman effect has been known for many years,[1] the resonant enhancement in the scattering cross section that occurs when the input frequency ω_1 is near an atomic absorption edge has been discovered only very recently by Sparks.[2] As Sparks correctly points out, a theoretical description of this phenomenon requires that the $\vec{p}\cdot\vec{A}$ term in the interaction Hamiltonian be taken to second order in perturbation theory. Sparks also notes correctly that the dispersion corrections to the atomic scattering factor[3] require a similar treatment, and he postulates a conservation law which he claims enables him to use the known form of these dispersion corrections to describe the resonance x-ray Raman effect. The justification for this postulated conservation law is claimed to be the existence of a similar law for the A^2 terms in the interaction Hamiltonian. Unfortunately, *neither* of these conservation laws exist,[4] and the theoretical expressions presented by Sparks[2] are inadequate.

We present here a theory of the resonant x-ray Raman effect which does not employ *ad hoc* postulates, but represents, instead, a direct evalua-

tion of second-order perturbation theory. Unlike Sparks's[2] expressions, which fortuitously agree with only a single datum point, our theory, when applied to Sparks's data,[2] correctly predicts both the integrated intensity and the detailed spectrum of the scattered radiation, as well as the surprising fact that the scattered radiation is completely unpolarized.

As is well known.[5] the $\vec{p}\cdot\vec{A}$ contribution to the differential cross section $d\sigma/(d\Omega\, d\omega)$ may be written

$$\frac{d\sigma}{d\Omega\, d\omega_2} = r_0^2\left(\frac{\omega_2}{\omega_1}\right)|M_{IF}|^2\delta\left[\frac{E_F - E_I}{\hbar} - (\omega_1 - \omega_2)\right], \tag{1}$$

where

$$M_{IF} = \sum_n[\langle I|\hat{u}_1\cdot\textstyle\sum_a \vec{p}_a|n\rangle\langle n|\hat{u}_2\cdot\sum_b \vec{p}_b|F\rangle/(\Omega_{nI} - \omega_1) + \langle I|\hat{u}_2\cdot\textstyle\sum_a \vec{p}_a|n\rangle\langle n|\hat{u}_1\cdot\sum_b \vec{p}_b|F\rangle/(\Omega_{nI} + \omega_2)]/m\hbar. \tag{2}$$

Here the sum on $|n\rangle$ is over all allowed intermediate states, a and b are electron labels, ω_2 is the frequency of the scattered photon, E_I (E_F) is the energy of the initial (final) state, $\Omega_{nI} = \hbar^{-1}(E_n - E_I)$, and the $\hat{u}$ are unit polarization vectors. In writing Eqs. (1) and (2) we have neglected the nonresonant A^2 contribution and have made the "dipole" approximation $e^{i\vec{k}\cdot\vec{r}} \to 1$. This latter is presently justified since for Cu $K\alpha$ radiation scattered by a Ni sample,[2] for example, $ka \sim 0.1$.

We evaluate Eq. (2) for a single isolated atom by assuming that the relevant many-electron wave function is adequately described by a single Slater determinant. In order to be specific, we treat in detail the case in which the final state contains an L-shell vacancy, since this case covers most of Sparks's data.[2] The intermediate states are of two kinds; those with a K-shell hole, and those with a hole in a higher (L, M, etc.) shell. In the former case the correct resonance condition may be seen to be

$$\omega_2 = \Omega_{K\alpha} \tag{3}$$

while for an intermediate L-shell hole, for example, the resonance condition is $\omega_2 = 0$. Here we take $\Omega_{K\alpha}$ to be the average frequency of the $K\alpha$ lines since we do not, at present, distinguish between the L_{II} and L_{III} levels. Because the conditions of Sparks's experiments[2] correspond to Eq. (3), we include only intermediate states with a K-shell hole.

For heavy elements it is usual, in the x-ray region,[3] to treat sums such as those in Eq. (2) by ignoring the separate existence of the empty atomic bound states since these are all clustered within a few electron volts of the continuum edge. Adopting this point of view, we have for the integrated intensity of the scattered radiation $d\sigma/d\Omega$

$$\frac{d\sigma}{d\Omega} = r_0^2 g_{1s,2p}\frac{\Omega_{K\alpha}}{\omega_1}(\Omega_{K\alpha} + \omega_1)^{-2}\int_0^{\omega_1 - \Omega_L} d\omega\left(\frac{dg}{d\omega}\right)_K \frac{(2\Omega_K - \Omega_L + \omega)^2(\Omega_K + \omega)(\omega_1 - \Omega_L - \omega)}{(\Omega_K - \omega_1 + \omega)^2}, \tag{4}$$

where Ω_K (Ω_L) is the K- (L-) shell ionization frequency, $g_{1s,2p}$ is the one-electron oscillator strength of a $1s \to 2p$ transition, $(dg/d\omega)_K$ is the one-electron oscillator density for transitions from the K shell to the continuum,[3] and where we have summed over all possible final-state configurations. Note that the polarization vectors $\hat{u}_1$ and $\hat{u}_2$ do not appear, so that the output is unpolarized. This surprising result is due to the two-electron nature of the scattering which may be described as absorption of ω_1 by a K-shell electron with simultaneous emission of ω_2 by an L-shell electron. When the resonance condition of Eq. (3) is satisfied, both these events correspond to real (energy-conserving) processes. We also note that the addition of electron-electron correlation will probably result in a small, but potentially important, degree of polarization.

Since we have neglected damping, the integral in Eq. (4) converges only if $\omega_1 < \Omega_K$. Sparks's experiments correspond to this case. For $\omega_1 \geq \Omega_K$, the scattered spectrum always has a nonintegrable singularity at $\omega_2 = \Omega_{K\alpha}$. In contrast to our

TABLE I. Cross section $d\sigma/d\Omega$ for the resonant x-ray Raman effect in units of r_0^2.

Element	Calc.[a]	Obs.[b]	Calc.[b]
Ni	6.94	7.0 ± 0.2	6.74
Cu	2.10	2.5 ± 0.4	3.4
Zn	1.17	1.3 ± 0.4	2.3
Ge	0.62	1.1 ± 0.4	1.6

[a]Our Eq. (4). [b]Ref. 2.

Eq. (4), Sparks's Eq. (4) (James[3]) implies that

$$\frac{d\sigma}{d\Omega} = 2r_0^2\left[\int_{\Omega_K}^{\infty} d\omega \left(\frac{dg}{d\omega}\right)_K \frac{\omega^2}{\omega_1^2-\omega^2}\right]^2 \text{(Sparks)}, \tag{5}$$

which is finite for all $\omega_1 \neq \Omega_K$.

In Table I we compare the predictions of Eqs. (4) and (5) with the experimental data of Sparks.[2] In making this comparison we use the atomic wave functions of Clementi[6] for computation of $g_{1s,2p}$, the well-known relationship between $dg/d\omega$ and the absorption coefficient,[3] and a widely accepted two-term approximation for the absorption coefficient.[7] Note that unlike Sparks's expressions which are in fortuitous agreement with experiment only for Ni, our theory correctly predicts the integrated intensity of the scattering.

The differences between the two theories are further clarified in Fig. 1 where we plot both Eqs. (4) and (5) versus $x = \omega_1/\Omega_K$. Although this graph is drawn for Ni, the other elements in Table I yield very similar results. From this figure we observe that for $x = 0.95$, the two curves cross, leading to a fortuitous agreement of Sparks's theory with experiment for Ni, while for smaller x (Cu, Zn, Ge) Sparks increasingly overestimates the cross section which, in his theory, fails to go continuously to zero, as it must, as $\omega_1 \to \Omega_L$. This qualitative relationship between Sparks's theory and his data is borne out by the entries in Table I. For Cu $K\beta$ radiation scattered from a Cu target, on the other hand ($x = 0.992$), Sparks's theory greatly underestimates the resonant enhancement of the x-ray Raman effect.

In Fig. 2 we plot our theoretical predictions for the spectrum $d\sigma/(d\Omega\, d\omega)$ of radiation scattered from a Ni target for different values of ω_1/Ω_K. Curve a corresponds to the conditions of Sparks's experiment, while curves b and c indicate the predicted form for $\omega_1 > \Omega_K$. Measurements of such spectra should provide a crucial test of the correctness of the theory presented here.

At present, the data available permit a test only of curve a in Fig. 2. In order to compare our predictions with Sparks's raw experimental data [his Fig. 1(b)] our theory must be normalized to the experimental peak and further modified to account for finite instrumental resolution, nonzero background, and internal absorption of the scattered radiation by the sample. The resulting comparison[8] is displayed in Fig. 3 where the very excellent agreement between our theory

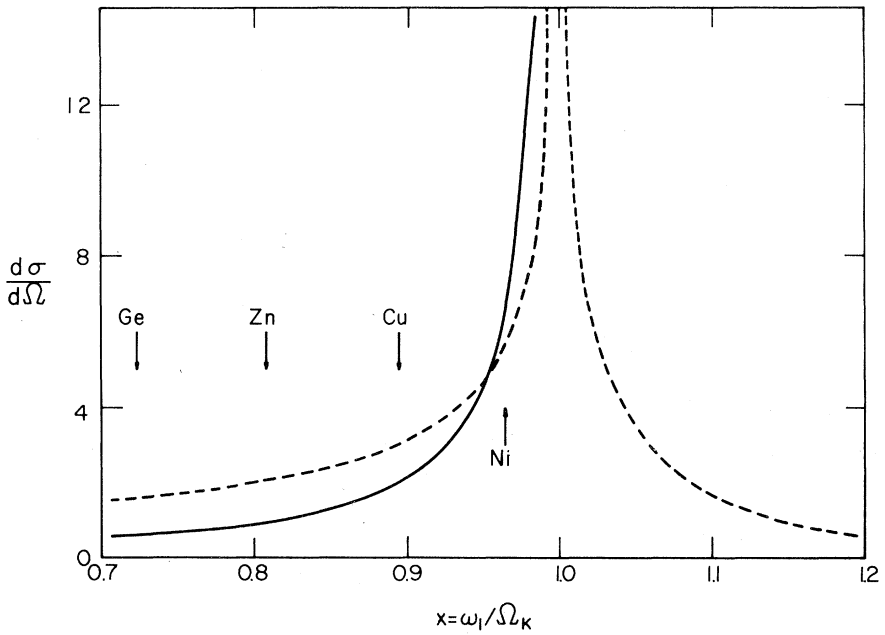

FIG. 1. The integrated intensity $d\sigma/d\Omega$ in units of r_0^2 of the resonant Raman x-ray effect. The solid line is our Eq. (4), the dashed line is Sparks's (Ref. 2) theory. Note that for Ge, Zn, and Cu Sparks overestimates the intensity, while for Ni there is fortuitous near agreement.

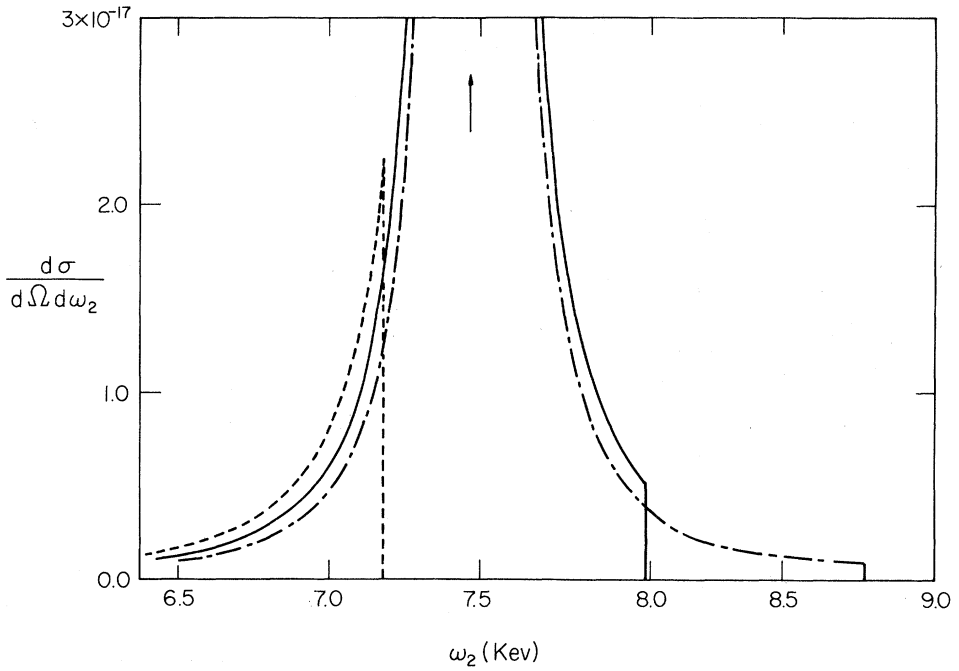

FIG. 2. The spectral density $d\sigma/d\Omega\, d\omega$ in units of r_0^2 sec for Ni for three different input frequencies, ω_1: (a) ----, $\omega_1 = \mathrm{Cu}\, K\alpha$; (b) ——, $\omega_1 = \mathrm{Cu}\, K\beta$; and (c) -•-•-, $\omega_1 = \mathrm{Au}\, L\alpha_2$. The arrow locates the pole in the spectrum that occurs at $\omega_2 = \Omega_{K\alpha}$ when $\omega_1 > \Omega_K$.

and Sparks's experiment is immediately apparent.

The resonant x-ray Raman effect is a new and potentially very important spectroscopic probe. As our analysis indicates, the scattered spectrum is expected to contain most of the information available from the absorption spectrum, such as the extended fine structure[9] and the absorption-edge singularities.[10] Because of the two-electron nature of the scattering, other specific effects due to electron-electron correlation are also anticipated. We note that there are many experimental advantages in measuring the x-ray Raman spectrum rather than the x-ray absorption spectrum—a similar situation exists in the infrared. We thus conclude that a theory more detailed than the one presented here, as well as extended high-resolution measurements, would be of great value in clarifying the potential of this new technique. Our efforts in this direction will be published elsewhere.

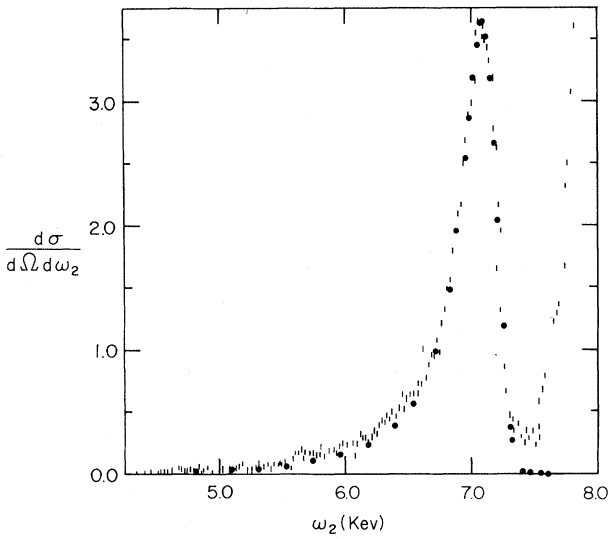

FIG. 3. Spectral density $d\sigma/d\Omega\, d\omega_2$ in arbitrary units for Cu $K\alpha$ radiation scattered from Ni. The ticks are Sparks's data (Ref. 2) from which a small constant background has been subtracted so as to cause the spectrum to asymptote to zero below 5 keV. The filled circles are our theoretical results modified to include instrumental broadening and sample absorption.

[1]K. Das Gupta, Phys. Rev. Lett. 3, 38 (1959); B. Davis and D. Mitchell, Phys. Rev. 32, 331 (1928).

[2]C. J. Sparks, Jr., Phys. Rev. Lett. 33, 262 (1974).

[3]R. W. James, *The Optical Principles of the Diffraction of X-Rays* (Cornell Univ. Press, Ithaca, N.Y., 1965), Chap. IV.

[4]The conservation law in question concerns the intensity of the total, coherent plus incoherent, scattering. For an atom with Z electrons, this intensity equals Z^2 for zero-scattering angle and falls to Z for large angle.

Only for $Z=1$ is the intensity "conserved."

[5]J. J. Sakurai, *Advanced Quantum Mechanics* (Addison-Wesley, Reading, Mass., 1967), Eq. 2.162.

[6]E. Clementi, IBM J. Res. Develop. 9, 2 (1965).

[7]*International Tables for X-Ray Crystallography*, edited by N. F. M. Henry and K. Lonsdale (Kynoch, Birmingham, England, 1965), Vol III, Sect. 3.2.

[8]We convolute our theoretical expression with a Gaussian instrumental function with full width at half-maximum (FWHM) of 197 eV appropriate to the 7-keV location of the peak and the 190 eV FWHM given by Sparks (Ref. 2) for an energy of 5.9 keV. The absorption of the sample is obtained from Ref. 7. Sparks's data were transferred directly using an opaque projector, and a small constant background correction was subtracted so as to cause the data to asymptote to zero (Ref. 2) below 5 keV. In order to bring our curve in register with Sparks's data, we found it necessary to translate our curve to lower energies by a very slight amount, ~ 27 eV. It is not presently known if this small discrepancy is an artifact due, perhaps, to calibration and drafting errors, or is real.

[9]D. E. Sayers, E. A. Stern, and F. W. Lytle, Phys. Rev. Lett. 27, 1204 (1971); F. W. Lytle, E. Sayers, and E. B. Moore, Jr., Appl. Phys. Lett. 24, 45 (1974).

[10]G. D. Mahan, Phys. Rev. 163, 612 (1967).