

Electron–Photon Coincidence Studies in Sodium

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Abstract

Three techniques are described from which the scattering parameters λ and χ can be measured in electron–photon coincidence studies of the 3^2P state of sodium. The relative sensitivity of the angular correlation and polarized photon techniques is discussed for states which have significant hyperfine structure in the excited state. The phenomenon of zero field quantum beats is described in the framework of the electron impact excitation of sodium. It is shown that the amplitude of the beat pattern depends on the parameter λ . The use of the beat pattern to determine λ is discussed.

1. Introduction

Over the past eight years electron–photon coincidence techniques have provided a very sensitive test of the theory of electron scattering from atoms. With these techniques it is possible to measure not only the magnitude but also the phase of the scattering amplitudes for discrete inelastic scattering from ground state atoms. Most of the experiments have concentrated on the targets hydrogen and helium, a fact which no doubt reflects the very high proportion of theoretical papers on such targets. The results of these experiments (see e.g. Weigold *et al.* 1980) on atomic hydrogen have shown that there is no theory which predicts accurately the magnitude and phase of the scattering amplitude in the intermediate energy range. The same conclusion can be drawn from the results for helium (see e.g. Hollywood *et al.* 1979).

Compared with hydrogen and helium there has been very little work done on sodium in the intermediate energy range either experimentally or theoretically. Indeed at 100 eV only two theories appear which describe the excitation of the 3^2P state. These are the Born approximation of D.L. Moores (personal communication, 1979) and a distorted wave polarized orbital (DWPO) calculation of Kennedy *et al.* (1977).

The differential cross section for the excitation of the 3^2P state of sodium has been measured by Buckman and Teubner (1979a) over a wide range of energies and scattering angles. We have found that, at 100 eV, both the Born approximation calculation and the DWPO calculation of Kennedy *et al.* (1977) accurately predict both the shape and absolute value of the differential cross section for scattering angles of $2^\circ \leq \theta_e \leq 18^\circ$. Indeed the differential cross section experiments cannot discriminate between these two theories in this angular range. For $\theta_e > 18^\circ$ the theories diverge rapidly from each other and from the experimental values. Although the DWPO calculation predicts qualitatively the shape of the differential cross section, it differs in absolute value by as much as a factor of ten for some scattering angles.

Thus, there seems little urgency to provide a more sensitive test of the theory of intermediate energy electron scattering from sodium at backward angles. Nevertheless the scattering parameters for angles less than 20° should be able to discriminate between the two theories at 100 eV.

For a P-S transition we use the formalism of Eminyian *et al.* (1974) to define the λ and χ parameters. Here the excited P state is described as a coherent superposition of eigenfunctions for each degenerate magnetic substate:

$$\psi(3^2\text{P}) = a_0 |1, 0\rangle + a_1 |1, 1\rangle + a_{-1} |1, -1\rangle,$$

where a_μ is the scattering amplitude for the excitation of the $m_L = \mu$ magnetic substate. The differential cross section for the excitation of this magnetic substate is

$$\sigma_\mu = \langle a_\mu a_\mu \rangle = |a_\mu|^2.$$

The reflection symmetry about the scattering plane ensures that $a_1 = -a_{-1}$ and thus

$$\sigma_1 = \sigma_{-1}.$$

The parameter λ is defined by

$$\lambda = \sigma_0 / (\sigma_0 + 2\sigma_1) = \sigma_0 / \sigma,$$

where σ is the total differential cross section for excitation of the P state.

We note that the assumption of reflection symmetry in the scattering plane implies that *LS* coupling may be used to describe the collision. We therefore also assume that exchange scattering processes can be ignored which is a reasonable assumption at 100 eV.

The scattering parameter χ measures the relative phase between the scattering amplitudes a_0 and a_1 :

$$\chi = \arg a_1 - \arg a_0.$$

There are several theories which relate the coincidence count rate to the parameters λ and χ and these will be discussed in more detail in Sections 2, 3 and 4. However, these theories have in general been evaluated only for states whose natural width is large compared with either fine or hyperfine structure. Such a simplification applies to helium and hydrogen but not to the resonant states of the alkalis and in particular the 3^2P state of sodium.

The most commonly occurring isotope of sodium, ^{23}Na , has nuclear spin $I = \frac{3}{2}$ and significant hyperfine structure (HFS) in both the $3^2\text{P}_{3/2}$ and $3^2\text{P}_{1/2}$ states. The lifetime of the 3^2P state is 16.3 ns which leads to a natural width of 61.3 MHz, comparable with the HFS splitting. It is this feature which not only produces special difficulties in coincidence experiments in sodium but is also responsible for the existence of modulation of the radiation emitted in the decay of the excited state.

In Section 2 the application of angular correlation experiments to sodium will be described; in Section 3 experiments are described in which the polarization of the decay photon is analysed prior to its detection in coincidence with the electron which has excited the state. In Section 4 we describe the phenomenon of zero field quantum beats in the electron impact excitation of the 3^2P state in sodium and discuss the influence of the parameter λ on the beat pattern.

2. Angular Correlation Techniques

The details of this technique as it applies to hydrogen and helium has been reviewed extensively by Blum and Kleinpoppen (1979). A typical geometry is shown in Fig. 1. Photons which are emitted at an angle θ_γ to the incident electron beam in the scattering plane are detected in coincidence with electrons which have excited the state and been scattered through an angle θ_e . The angular correlation is determined by measuring the number of true coincident counts as a function of θ_γ . A sinusoidal function is then fitted to the data to yield the scattering parameters λ and χ .

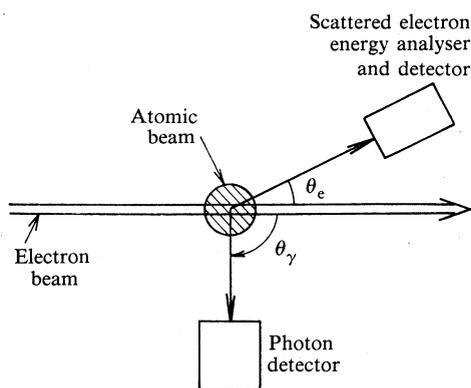


Fig. 1. Schematic diagram of the geometry employed in a typical angular correlation experiment.

The theory of angular correlations has been described by Macek and Jaecks (1971), who have shown that the coincidence count rate N can be described by

$$N = \text{const.} \times \sum_{m_L} \text{U} \langle a_{m_L} a_{m_L} \rangle \gamma \int_0^{\Delta t} \exp\{(-\gamma + i\omega)t\} dt.$$

The coefficients U contain all the angular momentum algebra for the problem and are given explicitly by equation (20) in Macek and Jaecks (1971). The scattering information is described by the density matrices $\langle a_{m_L} a_{m_L} \rangle$, whilst the time dependence of the data in the coincidence peak is contained in the integral. This last factor describes the modulation of the intensity in the beat pattern.

In the measurement of angular correlations one can take the upper limit $\Delta t \rightarrow \infty$ which yields

$$I = \gamma \int_0^{\infty} \exp\{(-\gamma + i\omega)t\} dt = \frac{\gamma^2}{\gamma^2 + \omega_i^2},$$

where $\omega_i = (E_{J'F'} - E_{JF})/\hbar$.

We note that for singlet P excitation in helium there is no fine or hyperfine structure and thus $I = 1$. For hydrogen we have $I \rightarrow 0$ for $J \neq J'$ and $I = 1$ for $J = J'$.

For sodium, I must be calculated for all hyperfine levels in each fine structure state. We have calculated I for the 3^2P state in sodium using the magnetic dipole moment constant a and the electric quadrupole moment constant b of Deech *et al.*

(1974). The resulting energy level diagram for sodium is shown in Fig. 2. The mean energy of the state and the fine structure splitting is taken from Moore (1971).

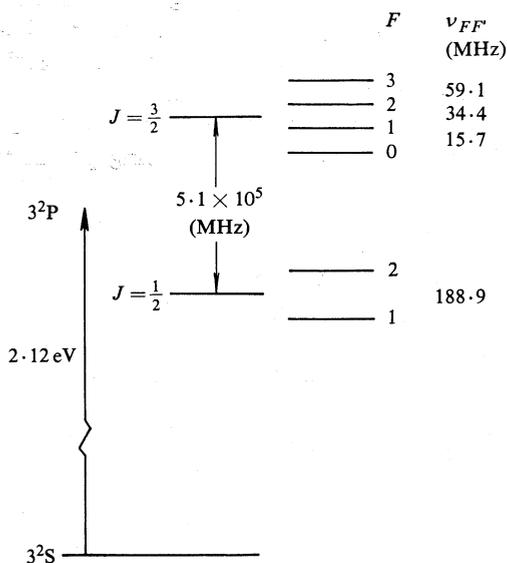


Fig. 2. Energy level diagram of the 3^2P state in sodium.

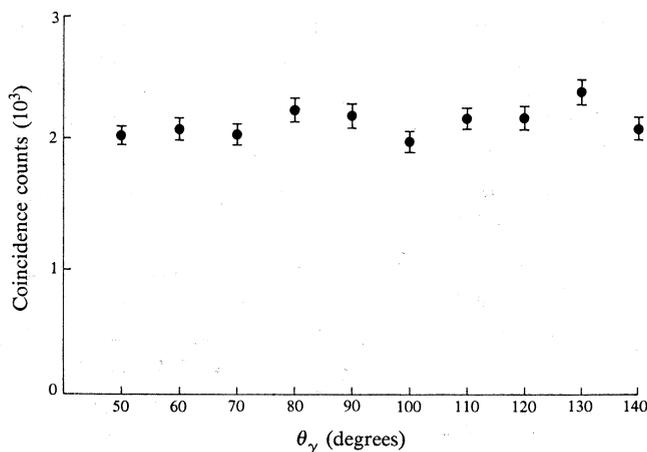


Fig. 3. Angular correlation for the excitation of the 3^2P state of sodium by 100 eV electrons; the electron scattering angle is 5° .

We have calculated the coefficients U for the 3^2P state in sodium and find that the coincidence count rate N for the excitation of this state is

$$N = C\sigma(0.65026 + 0.04923[\lambda + (1-\lambda)\cos 2\theta_\gamma - 2\lambda \cos^2\theta_\gamma + 2\{\lambda(1-\lambda)\}^{\frac{1}{2}}\cos \chi \sin 2\theta_\gamma])d\Omega_e d\Omega_\gamma, \quad (1)$$

where C is a constant. This angular correlation function is quite flat. Indeed the variation of the coincidence count rate with θ_γ is only about 15%.

An experimental angular correlation for the excitation of the 3^2P state in sodium by 100 eV electrons scattered through 5° is shown in Fig. 3. The data are consistent with $\lambda = 0$. This rather flat angular correlation underlines the limitations of this technique in the determination of the collision parameters. Thus it is profitable to investigate other techniques for the determination of the parameters λ and χ .

3. Polarized Photon Technique

The radiation arising from the decay of the 3^2P state to the ground state has wavelengths of 589 and 589.6 nm. Thus it is straightforward to analyse both the linear and circular polarization components of the radiation and therefore to measure all the components of the Stokes vector. These components are related to the scattering parameters λ and χ . The Stokes parameters P_i are defined by

$$IP_1 = I(0^\circ) - I(90^\circ), \quad IP_2 = I(45^\circ) - I(135^\circ), \quad IP_3 = I(\text{RHC}) - I(\text{LHC}),$$

where I is the total intensity of the radiation, $I(\alpha)$ is the intensity of the radiation when the optic axis of the analyser is at an angle α to the incident electron beam direction, and $I(\text{RHC})$ and $I(\text{LHC})$ are the intensities of the right- and left-handed circularly polarized radiation respectively. A typical geometry is shown in Fig. 4. The photons arising from the decay of the atomic state are analysed with the polaroid filter. After analysis they are counted in coincidence with the electrons which excited the state. The Stokes parameters are then determined by the variation of the true coincidence count rate with the optic axis of the analyser.

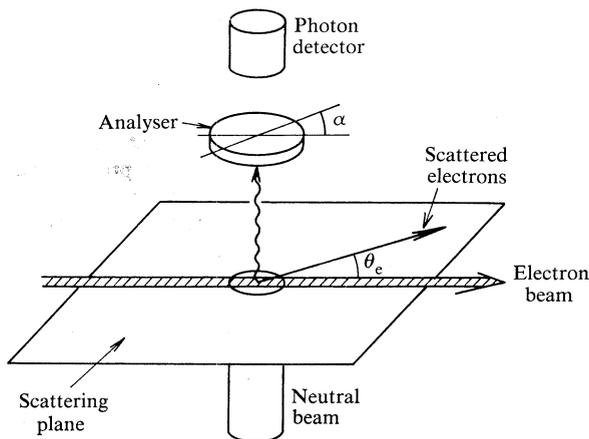


Fig. 4. Schematic diagram of a typical geometry used in the polarized photon technique.

This technique has been used in electron scattering by Standage and Kleinpoppen (1976) to study the excitation of the 3^1P state in helium and in atom-atom collisions by Kempter and collaborators (Zehnle *et al.* 1978; Menner *et al.* 1981).

For states with unresolved HFS, the relationship between the collision parameters and the components of the Stokes vector has been derived by Fano and Macek (1973). Adopting the formalism of Zehnle *et al.* (1978), we have the relationships

$$P_1 = \frac{3h^{(2)}G^{(2)}}{4-h^{(2)}G^{(2)}}(1-2\lambda),$$

$$P_2 = \frac{6h^{(2)}G^{(2)}}{4-h^{(2)}G^{(2)}}\{\lambda(1-\lambda)\}^{\frac{1}{2}}\cos\chi,$$

$$P_3 = -\frac{6h^{(1)}G^{(1)}}{4-h^{(2)}G^{(2)}}\{\lambda(1-\lambda)\}^{\frac{1}{2}}\sin\chi,$$

where the coefficients $h^{(k)}$ and $G^{(k)}$ are given by equations (8) and (40) in Fano and Macek (1973).

We find* for sodium that (Furst *et al.* 1982)

$$P_1 = 0.141(2\lambda - 1), \quad (2a)$$

$$P_2 = -0.282\{\lambda(1-\lambda)\}^{\frac{1}{2}}\cos\chi, \quad (2b)$$

$$P_3 = -1.116\{\lambda(1-\lambda)\}^{\frac{1}{2}}\sin\chi. \quad (2c)$$

The measurement of all three Stokes parameters will completely determine λ and the absolute value of χ .

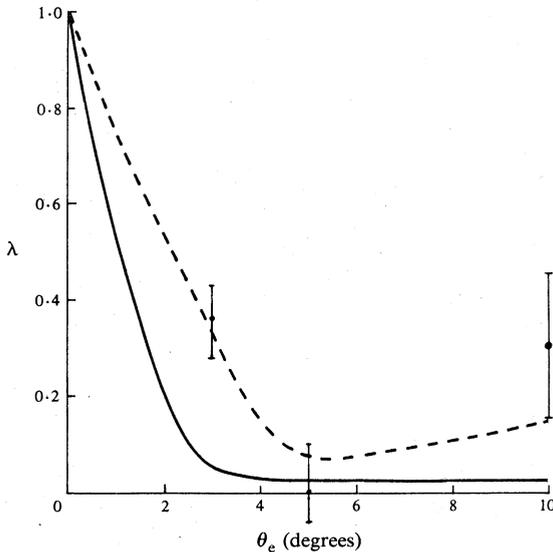


Fig. 5. Parameter λ as a function of the electron scattering angle θ_e at an incident energy of 100 eV: experimental points, present results; solid curve, Born approximation ($\lambda = \cos^2\theta_e$); and dashed curve, DWPO calculation.

Equations (2) exhibit an influence of the HFS on the polarization of the emitted radiation which is the same as that demonstrated in the analysis of total polarization by Flower and Seaton (1967). One consequence of such depolarization effects is that the variation of P_1 is only between ± 0.14 . Thus the constraints on the measurement of λ by this technique are such that very long counting times are required even at relatively small scattering angles. For example, the value of $\lambda = 0.30 \pm 0.15$ at $\theta = 10^\circ$ arose from 130 hours running time.

* These expressions differ from those used in our earlier work (Buckman and Teubner 1979b). The difference arose because of arithmetical mistakes in the calculation of $G^{(1)}$ and $G^{(2)}$.

Equation (2a) has been used to determine λ for several scattering angles for an incident energy of 100 eV. The results for sodium are shown in Fig. 5 and, even with the limited experimental accuracy of the data, they clearly discriminate against the first Born approximation.

4. Zero Field Quantum Beats

The HFS in the 3^2P state of sodium which causes problems in both the angular correlation and polarized photon techniques is responsible for another phenomenon, the analysis of which can yield the parameter λ . This phenomenon is known in beam foil spectroscopy as zero field quantum beats. Macek and Jaecks (1971) showed that in atomic states where the splitting is comparable with the natural width, the emitted light will be modulated due to the interference of radiation from the coherently excited levels of a hyperfine multiplet. This feature has been confirmed in beam foil spectroscopy (Andrä 1974) but has previously been ignored in electron-photon coincidence experiments. The topic has been discussed theoretically in relation to coincidence experiments (see e.g. Blum and Kleinpoppen 1979; Blum 1980). However, the discussion has centred on fine structure beats in atomic hydrogen which are impossible to observe with current technology. Sodium on the other hand appears a likely candidate for producing beats in the 3^2P state. The energy levels in Fig. 2 show that the HFS splitting in this state is of the order of the natural width of the level.

The theory of Macek and Jaecks (1971) has been used to find an expression for the coincidence count rate in an electron-photon coincidence experiment involving the 3^2P state in sodium. The photons were detected at $\theta_\gamma = 90^\circ$ in the scattering plane without regard to their polarization, and thus we have summed over two polarization directions β and $\beta + \frac{1}{2}\pi$. The coincidence count rate N is given by

$$N = K(A_{00} + A_{11} + A_{1-1}), \tag{3}$$

where

$$A_{qq'} = \sum_{JJ'FF'M_L M'_L} U(qq'JJ'FF'M_L M'_L) \langle a_{M_L} a_{M'_L} \rangle \int_0^{\Delta t} \exp\{-(\gamma + i\omega)t\} dt. \tag{4}$$

The coefficients U have been calculated using equation (20) in Macek and Jaecks (1971).

We have assumed that LS coupling can be used to describe the collision; thus, there is symmetry about the scattering plane and $a_1 = -a_{-1}$. In addition we assume that LS coupling may be used to describe the decay.

We note that there is some ambiguity about the limits of integration in the integral in equation (4). In the original formulation Macek and Jaecks stated that Δt is the resolution time of the circuits. This is equivalent to using an instrumental function of width Δt and is therefore only valid for the case $\Delta t \ll \tau$. A recent modification has been made by Parker *et al.* (1982) who took the limits from $-\frac{1}{2}\Delta t$ to $+\frac{1}{2}\Delta t$. Both of these cases however underestimate the influence of the timing resolution on the beat pattern.

In order to take account of the timing resolution we have replaced the time integral in (4) with the expression

$$f(t') = \int_{-\infty}^{\infty} P(t-t') \exp\{(-\gamma + i\omega)t'\} dt,$$

where $P(t-t')$ is a gaussian apparatus function. The integration over $\exp(i\omega t)$ introduces phase factors ϕ_i into the time dependence of the beat pattern. Work is still in progress on the explicit form of these phase factors.

Equation (3) can be expressed in the form

$$N = \exp(-\Gamma t) \left(A + B \sum_i \beta_i \cos(\omega_i t + \phi_i) \right),$$

where $\Gamma t = -\frac{1}{2}S^2\gamma^2 + \gamma(t-t_0)$ and S is the standard deviation of the gaussian apparatus function and t_0 the prompt response time of the circuits.

The angular frequencies are in this case given by

$$\omega_i = (E_{J'F'} - E_{JF})/\hbar,$$

and the coefficients β_i are determined by the constants U in equation (4) modified by the timing resolution.

It can be shown that

$$B = (3\lambda - 2) \times 10^{-2}.$$

In other words the relative amplitude of the beat pattern depends upon the value of λ .

Of the many possible combinations in the HFS levels, the angular momentum algebra restricts only those in the $J = \frac{3}{2}$ level. Specifically only the states with $FF' = 32, 21, 31$ and 20 contribute to the beats. The angular frequencies $\omega_{FF'}$ associated with these contributions are given in Table 1.

Table 1. Angular frequencies of components to HFS beats

| FF' | 32 | 21 | 31 | 20 |
|--|------|------|------|------|
| $\omega_{FF'} (10^8 \text{ rad s}^{-1})$ | 3.72 | 2.23 | 5.93 | 3.26 |

We have recently observed zero field quantum beats in the electron impact excitation of the 3^2P state of sodium (Teubner *et al.* 1981). A schematic diagram of the apparatus is shown in Fig. 6. A beam of sodium atoms was formed in a monel oven and entered the interaction region where it was crossed with a well focussed 100 eV electron beam. A cylindrical mirror electron spectrometer viewed the interaction region and analysed the energy of the electrons which had been scattered through 3° . Electrons which had excited the 3^2P state of sodium and therefore had lost 2.1 eV of energy were detected in a channel electron multiplier. Pulses from this detector were amplified and processed with standard timing electronics (CFTD) and acted as the start pulse for a time-to-amplitude converter (TAC).

The 589 and 589.6 nm photons arising from the decay of the 3^2P state passed through an interference filter of bandwidth 10 nm and were detected in a fast low noise photomultiplier tube (PMT: EMI 9863B). Pulses from the tube were amplified, delayed and processed by timing electronics similar to the electron channel and acted as the stop pulse for the TAC. The output of the TAC was recorded in a multi-channel analyser (MCA).

A typical timing spectrum is shown in Fig. 7. The lifetime of the 3^2P state has been determined from these data using the technique of Imhof and Read (1977). The data have been fitted to a function of the form

$$I(t) = b + a \operatorname{erf} \left\{ s^{-1} \sqrt{\frac{1}{2}}(t-t_0) - \sqrt{\frac{1}{2}} s \gamma \right\} \exp \left\{ \frac{1}{2} s^2 \gamma^2 - \gamma(t-t_0) \right\}, \quad (5)$$

where b is the height of the random background, t_0 the prompt response time, s the standard deviation of the gaussian instrumental response, $2a$ the peak height and γ the transition probability of the state. The curve through the data in Fig. 7 is a function of this form. The fit shows that the timing resolution of the system is 9.4 ns (FWHM) and the lifetime of the state is 16.3 ± 0.15 ns. The lifetime is in excellent agreement with the preferred value of the oscillator strength given by Fischer (1976).

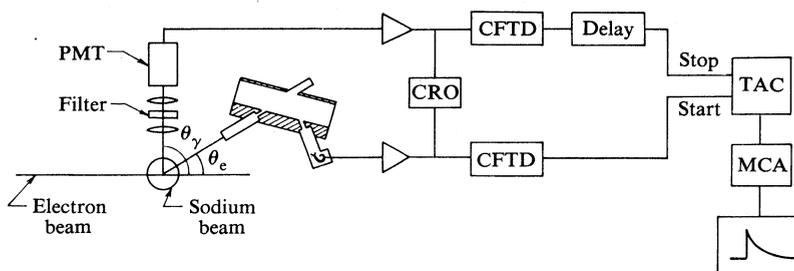


Fig. 6. Schematic diagram of the apparatus used in quantum beat studies in sodium.

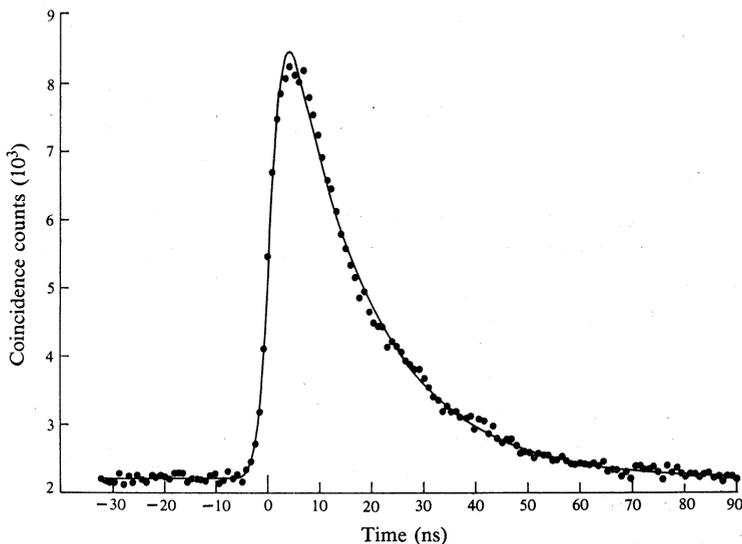


Fig. 7. Timing spectrum showing the decay of the 3^2P state in sodium. The curve represents a fit to the data of the form of equation (5).

The beats can be seen clearly as an oscillation of the data about the fit. The beat pattern is shown in Fig. 8. Here the pattern has been fitted to a function of the form

$$f(t) = a + \sum_i b_i \cos(\omega_i t + \phi_i).$$

The frequencies ω_i are as given in Table 1, but the coefficients b_i and ϕ_i have been allowed to vary freely. In principle b_i and ϕ_i are fixed by the angular momenta of the problem and by the experimental conditions, so that the only free parameter

in the fit is λ . The statistical accuracy of the data and the marginal timing resolution hampers the determination of λ from these data. We can however make the qualitative observation that the ease of observing the beat pattern follows the variation of λ with θ_e as shown in Fig. 5.

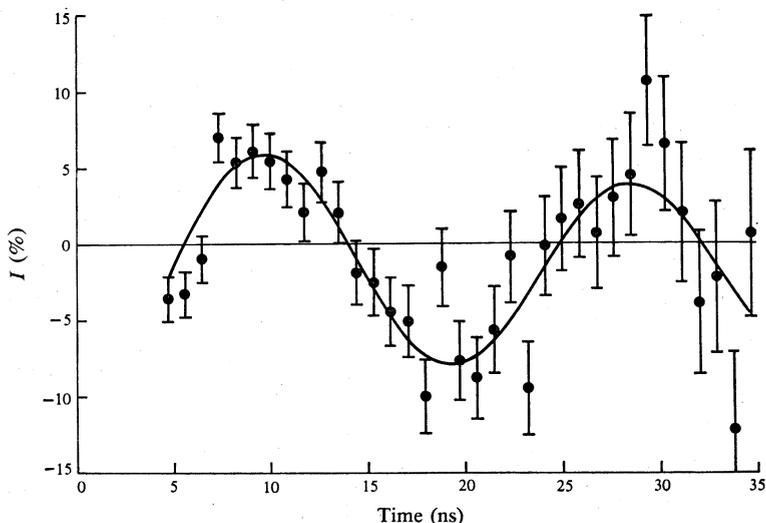


Fig. 8. Fit of the beat pattern to a sinusoidal function.

5. Conclusions

The presence of significant HFS in the first excited state of sodium causes specific difficulties in the measurement of the scattering parameters for this state. Nevertheless, we have shown that it is possible to measure λ with sufficient accuracy at 100 eV so that one can distinguish between the two theories even at forward angles. The rather poor state of the theory at backward angles has not improved since the work of Kennedy *et al.* (1977). This is somewhat surprising when one considers the dominant contribution which the 3^2P state makes to the total oscillator strength for the collision. Thus it is extremely likely that a 'few state' close coupling calculation will provide accurate answers even at 100 eV. We await with interest the application of the coupled channel optical model of Stelbovics and McCarthy (1982) to this problem.

The techniques which we have described above can be applied at lower incident energies. It will be particularly relevant to measure the scattering parameters at 10 and 20 eV where we can compare results with the super-elastic scattering experiments of Hertel and collaborators (Hermann and Hertel 1982). These experiments are the time and parity inverse of the coincidence experiments described in this paper.

Acknowledgments

We gratefully acknowledge discussions with S. J. Buckman. This research was supported in part by a grant from the Australian Research Grants Scheme. One of us (J.L.R.) acknowledges the assistance of a Commonwealth Post Graduate Research Award.

References

- Andrä, H. J. (1974). *Phys. Scr.* **9**, 257.
- Blum, K. (1980). In 'Electronic and Atomic Collisions' (Eds N. Oda and K. Takayanagi), p. 579 (North-Holland: Amsterdam).
- Blum, K., and Kleinpoppen, H. (1979). *Phys. Rep.* **52**, 203.
- Buckman, S. J., and Teubner, P. J. O. (1979a). *J. Phys. B* **12**, 1741.
- Buckman, S. J., and Teubner, P. J. O. (1979b). *J. Phys. B* **12**, L583.
- Deech, J. S., Hannaford, P., and Series, G. W. (1974). *J. Phys. B* **7**, 1131.
- Eminyan, M., MacAdam, K. B., Slevin, J., and Kleinpoppen, H. (1974). *J. Phys. B* **7**, 1519.
- Fano, U., and Macek, J. (1973). *Rev. Mod. Phys.* **45**, 553.
- Fischer, C. F. (1976). *Can. J. Phys.* **54**, 1465.
- Flower, D. R., and Seaton, M. J. (1967). *Proc. Phys. Soc. London* **91**, 59.
- Furst, J. E., Riley, J. L., Teubner, P. J. O., and Buckman, S. J. (1982). *J. Phys. B* (to be published).
- Hermann, H. W., and Hertel, I. V. (1982). *Comments At. Mol. Phys.* **12**, 61.
- Hollywood, M. T., Crowe, A., and Williams, J. F. (1979). *J. Phys. B* **12**, 819.
- Imhof, R. E., and Read, F. H. (1977). *Rep. Prog. Phys.* **40**, 3.
- Kennedy, J. V., Myerscough, V. P., and McDowell, M. R. C. (1977). *J. Phys. B* **10**, 3759.
- McCarthy, I. E., and Stelbovics, A. (1982). *Aust. J. Phys.* **35**, 543.
- Macek, J., and Jaecks, D. H. (1971). *Phys. Rev. A* **4**, 2288.
- Menner, B., Hall, Th., Zehnle, L., and Kempster, V. (1981). *J. Phys. B* **14**, 3693.
- Moore, C. E. (1971). 'Atomic Energy Levels', NSRDS-35, Vol. 1 (National Bureau of Standards: Washington D.C.).
- Parker, G. A., Miller, T. M., and Golden, D. E. (1982). *Phys. Rev. A* **25**, 588.
- Standage, M. C., and Kleinpoppen, H. (1976). *Phys. Rev. Lett.* **36**, 577.
- Teubner, P. J. O., Furst, J. E., and Riley, J. L. (1982). to be published.
- Teubner, P. J. O., Furst, J. E., Tonkin, M. C., and Buckman, S. J. (1981). *Phys. Rev. Lett.* **46**, 1569.
- Weigold, E., Frost, L., and Nygaard, K. G. (1980). *Phys. Rev. A* **21**, 1950.
- Zehnle, L., Clemens, E., Martin, P. J., Schäuble, W., and Kempster, V. (1978). *J. Phys. B* **11**, 2865.

