Physica A 157 (1989) 395–399
North-Holland, Amsterdam

FAR INFRARED ABSORPTION BY SMALL BISMUTH PARTICLES

R.E. SHERRIFF and R.P. DEVATY

Department of Physics and Astronomy, 100 Allen Hall, University of Pittsburgh, Pittsburgh, PA 15260, USA

We have observed low frequency (<50 cm^{-1}) magnetic field dependent resonances and higher frequency resonances at zero field in the far infrared (FIR) absorption of small bismuth particles. A model based on the quasistatic approximation shows both good agreement with previous measurements of resonance frequencies vs. field and qualitative agreement with our data. Additional work is required to quantify the comparison.

Keywords: Infrared spectra, Absorption, CPH cs, Bismuth, Resonances, Reprints. (JG)

1. Introduction

Bismuth is often used as a model system for solid state plasmas because many of its important energies (E_F, $\hbar\omega_p$, etc.) are conveniently close together in the FIR. The properties of Bi make it especially interesting for FIR magneto-optical studies because the interaction of the cyclotron-like resonances with the plasma modes can be seen using currently achievable fields. The low carrier density also implies a large Kubo gap [1], enhancing the possibility of seeing quantum size effects (QSE) or metal–insulator transitions.

One of the earliest studies of small Bi particles was carried out by Chin and Sievers [2]. Motivated by the idea that free Bi particles appeared to align the bisectrix axis along the applied magnetic field [3], they performed FIR transmission measurements on free-standing Bi particles and found three field dependent resonances. No successful explanation of these lines was given. We have developed a model using bulk Bi parameters that agrees well with the data. The model also predicts additional resonances in and above the frequency range covered by Chin. To search for these additional resonances, we investigated the FIR absorption of small Bi particles both at low frequencies in a magnetic field and at higher frequencies at zero field.

2. Theory

Chin's data (fig. 1) can be successfully modelled by treating the Bi particles as small spheres with an anisotropic dielectric tensor $\mathscr{E}$. The quasistatic

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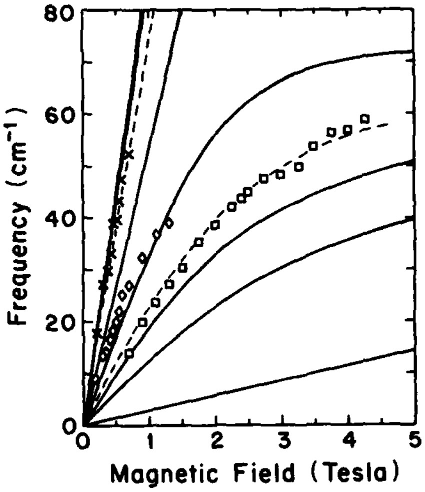

Fig. 1. Field dependent resonances for 4000 Å diameter free standing bismuth particles. The symbols show data while the lines represent calculations based on our model. Solid lines indicate magnetic dipole curves while dashed lines are electric dipole.

boundary value problem [4] is solved to find α (the absorption coefficient),

$$\alpha(\omega) = \frac{9}{2} f\sqrt{\epsilon_0}\left(\frac{\omega}{c}\right) \mathrm{Im}\{\hat{E}^* \cdot \boldsymbol{B}^{-1} \cdot \hat{E}\} + \frac{1}{5} f d^2 \sqrt{\epsilon_0}\left(\frac{\omega}{c}\right)^3 \mathrm{Im}\{\hat{B}^* \cdot \overline{\mathscr{E}^{-1}}^{-1} \cdot \hat{B}\}\,, \tag{1}$$

where ω is the angular frequency, ϵ_0 the dielectric constant of the surrounding medium, $\mathscr{E}$ the dielectric tensor of the sphere, d the particle diameter, and f the volume fraction of Bi. The tensors in the expression are given by $\boldsymbol{B} = \boldsymbol{1} + 2\epsilon_0 \mathscr{E}^{-1}$ and $\overline{\mathscr{E}^{-1}} = \mathrm{Tr}(\mathscr{E}^{-1})\boldsymbol{1} - (\mathscr{E}^{-1})^{\mathrm{T}}$.

To get $\mathscr{E}$ for a small Bi sphere, we use an anisotropic Drude model with bulk parameters [5] for the four carriers:

$$\mathscr{E} = \mathscr{E}_{\mathrm{L}} - \frac{4\pi q^2}{\omega\bar{\omega}} \sum_{j=1}^{4} n_j (\boldsymbol{m}_j^*)^{-1}\,. \tag{2}$$

Here n_j is the number density and $\boldsymbol{m}_j^*$ the effective mass tensor for the jth carrier. $\mathscr{E}_{\mathrm{L}}$ is a frequency independent tensor [5], q the electronic charge, and $\bar{\omega} = \omega + \mathrm{i}/\tau$. [Note: The magnetic dipole term (2nd term in eq. (1)) is incorrect due to neglect of the electric quadrupole term [6]. However, the large value of $\mathscr{E}_{\mathrm{L}}$ for Bi makes the corrections small.]

Fig. 1 compares the Chin data with the theory. The data agree remarkably well with three of the theoretical curves, allowing us to identify two of the curves as electric dipole resonances, and one as magnetic dipole. The model, however, also predicts a large number of lines that are not seen in these data as well as resonances that should exist at higher frequencies, even with $H = 0$.

3. Experiment

We used two approaches to test the model. First we looked for additional low frequency field dependent resonances in free-standing powders. The powders themselves limited the upper cutoff frequency to around 50 cm^{-1}. To search for higher frequency resonances we used pressed pellets of Bi in paraffin. This enabled us to use low volume fractions of Bi and raise the cut-off frequency.

The Bi particles were prepared by inert gas evaporation in Ar [7]. Size distributions were obtained from transmission electron micrographs. Electron diffraction patterns indicated that the particles were crystalline.

Samples of free-standing powders were made by spreading a thin, approximately uniform layer of powder onto a Mylar sheet and then sandwiching it with another layer of Mylar to keep it in place during sample insertion.

To prepare pressed pellets, the Bi powder was first mixed with paraffin powder and manually shaken. The paraffin was then melted and the suspension of Bi particles was stirred. After resolidifying, the sample was repeatedly ground in a freezer mill at 77 K and then pressed into half inch diameter pellets. The grinding/pressing cycle was repeated 3–4 times.

The transmission of the free-standing powder was measured in the Faraday geometry for various magnetic fields using a Michelson interferometer over the range 10–50 cm^{-1}. For the pressed pellets, we measured the FIR absorption coefficient, again with a Michelson but over the range 30–300 cm^{-1}.

The results of our study on a free-standing Bi powder appear in fig. 2 as ratios of spectra with field to spectra without field. Arrows indicate the locations of possible resonances, but many of the identifications must be considered tentative because of their small size (slightly larger than noise in the spectra). Even discounting some of the smaller, less trustworthy resonances, the data still show an abundance of field dependent absorption peaks providing at least qualitative support for our model.

Quantitative comparison on the other hand is difficult. In addition to the small size of many resonances, interpretation of the data is further complicated by the changing size of the peaks as they move upward in frequency. This makes it difficult to unambiguously trace the field dependence of each resonance from curve to curve, especially in regions where the magnetic field

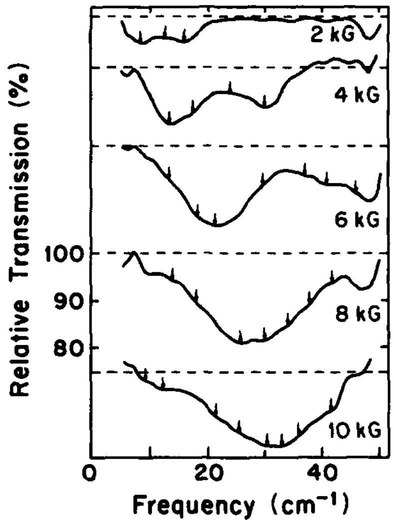

Fig. 2. Relative transmission spectra of 5000 Å diameter free standing bismuth particles for five different values of the magnetic field. The dashed lines are at the 100% level for each curve. All the curves are plotted with the same vertical scale. Arrows indicate possible locations of resonance peaks.

sampling interval is more than 1 kG. Further confusion results from the low transmission level of the samples which both decreases SNR (raising doubts about the smaller resonances) and limits the frequency range over which resonances can be tracked. A direct quantitative evaluation of the model beyond the agreement with Chin's data must wait until additional experiments in progress are completed.

It is possible however to obtain support for our model from the pressed pellet data shown in fig. 3. The absorption coefficient at zero field shows a resonance within 7% of the predicted location for the zero field combined dipole peak for randomly oriented particles. The theory predicts two peaks but it is possible that the resonances are too broad to be resolved.

4. Conclusions

We have developed a model for the far infrared absorption of small Bi particles that agrees with the data of Chin [2]. Our own experimental evidence of a large number of field dependent resonances below $50\,cm^{-1}$ and a strong resonance around $180\,cm^{-1}$ at zero field also suggests at least a qualitative agreement with the model. A direct quantitative comparison awaits the collection of more data.

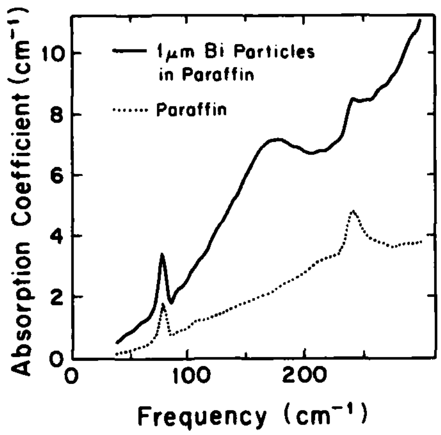

Fig. 3. FIR absorption coefficient for 1 μm bismuth particles in paraffin. The absorption coefficient for paraffin is shown for comparison.

Acknowledgement

This work was supported by ONR Contract #N00014-85-K-0808.

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